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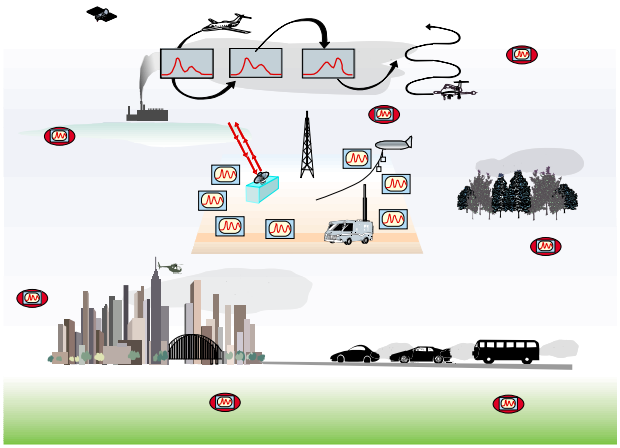
Tropospheric Aerosol Program



Program Plan

March 2001

U. S. Department of Energy
Office of Science
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Foreword

The Department of Energy (DOE) and its predecessor agencies, the Atomic Energy Commission and the Energy Research and Development Administration, have a long and enviable record of accomplishment in the science of atmospheric aerosols. This research, which had its genesis in the study of fall-out from atmospheric weapons testing, has found valuable new application in understanding the environmental effects of fossil fuel combustion and allied energy-related activities.

Now, consistent with the Nation's desire to preserve and enhance our environment and minimize the risk to human health and welfare from atmospheric pollutants, the atmospheric science research community faces a new challenge to develop sensible and effective strategies to achieve the new National Ambient Air Quality Standard for fine particles, the so-called PM-2.5 standard. Achieving this standard in a way that will have minimum impact on the Nation's ability to meet its energy requirements requires a much more complete understanding of the processes governing the loading, composition, and microphysical properties of these aerosols than is now available.

Fine particles are implicated in another important issue that may affect the Nation's energy economy, namely climate change. Fine particles scatter solar radiation, decreasing the amount of the sun's energy that is absorbed by the planet and thereby exerting a cooling influence on climate. The magnitude of this influence is not known for certain, but recent estimates indicate that it is comparable to the warming influence of increased concentrations of greenhouse gases and may consequently be offsetting a major fraction of the greenhouse warming that would otherwise have been experienced over the industrial period. Because aerosols are short-lived in the atmosphere this effect cannot be considered a mechanism for forestalling the greenhouse effect. But to understand climate change it is necessary to obtain accurate estimates of the totality of climate influences over the industrial period, and in particular the aerosol influences.

The Tropospheric Aerosol Program (TAP) described in this Program Plan will make crucially-needed contributions to improved understanding and model-based description of the loading and properties of atmospheric aerosols in relation to sources, pertinent to both of these major environmental issues. Scientists from the DOE National Laboratory community together with colleagues from the academic community, the private sector, and other governmental agencies responsible for understanding and maintaining our atmospheric environment have contributed substantially to the preparation of this Plan.

The talent required to understand and resolve these important national issues lies collectively within and beyond the Department of Energy. Thus we view TAP as a component of a larger, informal national aerosol program, where TAP both contributes to and leverages other aerosol research efforts. Indeed, TAP is designed to fill some very important gaps and complements existing programs. We look forward to working closely with our partners within DOE and in other state and federal agencies, industry, and academia.

With this cooperative effort TAP will serve the objectives of these communities and thereby make a major contribution to meeting the goals of the Air Quality Research Subcommittee of the Committee on Environment and Natural Resources, while at the same time supporting the DOE mission of fostering a National Energy Strategy that takes into account the preservation and enhancement of the Nation's atmospheric environment.

Dr. Ari Patrinos
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Environmental Research
Office of Science
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Executive Summary

Air quality in the United States has improved substantially over the past three decades as a result of the Clean Air Act and its amendments. However, a few major unresolved issues remain in accomplishing the task of minimizing the public's risk to air pollution exposure. Key among these is the relation of adverse health effects to exposure to airborne particles, or "aerosols".

The significance of the environmental stress of particulate air pollution and the mechanisms of its action on humans remain unresolved despite years of research. Historically, attention has shifted from crude measures of exposure represented by the mass concentration of "total suspended particles" to more refined measures that segregate mass concentration by the upper limit particle size, initially 10 μm and more recently 2.5 μm. However it is not established that it is the mass concentration of particulate matter that is in fact responsible for its health effects. As knowledge has evolved and measurement methods have improved, health scientists have begun to turn their attention to the chemical components present in particles. This has led to the recognition that available means of determining chemical properties may be limiting the identification of components in particles responsible for their influence on human health.

In addition to their influence on health, fine particles affect public welfare in other important ways. These particles scatter and absorb light, leading to impairment of visibility and to a potential influence on climate change that must be quantified to understand the full implications of increasing concentrations of greenhouse gases. Light scattering and absorption by fine particles also affect surface irradiance, with a potential resultant influence on plant growth and length of the growing season. Deposition of fine particles and their precursors to the surface affects the chemical balance of ecosystems, not just by so-called "acid deposition" but also by alteration of nutrient balance. For these reasons also quantitative understanding is required of the processes that control the atmospheric loading, composition, and microphysical properties of fine particles, together with the capability to represent these processes in atmospheric chemical transport models.

In 1997, the U.S. government made a commitment to reduce fine particles in the air by modifying the National Ambient Air Quality Standards (NAAQS) by establishing a new standard for fine particulate matter, the PM2.5 standard. The public debates that took place in developing this standard revealed that scientific information supporting this national objective continues to have unacceptably high levels of uncertainty. As a result, the nation has embarked on a substantial new program of research to reduce these uncertainties. Initial attention has focused mainly on two of the technical issues necessary to reduce this uncertainty: characterizing human health effects due to exposure to fine particles and determining the nature and extent of fine particle exposure across the United States, as specified by the new NAAQS.

While the above two elements are key to defining the root cause of the health effects and to identifying the geographic locations of non-attainment, they will fall short of leading to an efficient approach to controlling the problem. To achieve this goal the new national program needs two more elements: a greatly improved system for quantitatively characterizing emissions, and greatly improved understanding of the basic physico-chemical processes responsible for the formation of particles and evolution of their properties in the atmosphere. Without these elements it will not be possible to link emission reductions with reductions in particle concentrations and thereby formulate a rational and efficient strategy to achieve the PM2.5 standard. Absence of this knowledge also precludes developing strategies that would be targeted to specific substances in fine particulate matter that might achieve real benefits to human health.

The uncertainties in specifying emissions and atmospheric processes linking sources and atmospheric concentrations are as numerous and complex as those linking health effects to exposures. Thus, a substantial investment in process focused research is needed for decision-makers to ensure that the best information is available to construct new strategies for minimizing the health and environmental consequences of exposure to fine particles resulting from energy production or use. Improved emissions inventories are now being developed by the air pollution research community under support from a variety of sources. However the study of the atmospheric processes that govern the evolution of mass loading and chemical and physical properties of aerosols remains a weak link in the chain.

The Department of Energy's Tropospheric Aerosol Program (TAP) will, in collaboration with research by NOAA, EPA, and other Federal and state agencies, address the fourth critical element, the atmospheric science of fine particles. TAP falls within the purview of the Department of Energy for a number of reasons. Fine particles in the atmosphere derive largely from energy production and use; DOE has a statutory responsibility to conduct research into the health and environmental consequences of these activities. Moreover, any control measures that are proposed will likely have considerable impacts on energy production and consumption technologies and energy costs. DOE also has a long history of distinguished research in atmospheric aerosols, photochemistry, and meteorology and as a consequence has the requisite pool of talent to examine these complicated processes in detail.

The goal of TAP will be to develop the fundamental scientific understanding required to construct tools for simulating the life cycle of tropospheric aerosols--the processes controlling their mass loading, composition, and microphysical properties, all as a function of time, location, and altitude. The TAP approach to achieving this goal will be by conducting closely linked field, modeling, laboratory, and theoretical studies focused on the processes controlling formation, growth, transport, and deposition of tropospheric aerosols. This understanding will be represented in models suitable for describing these processes on a variety of geographical scales; evaluation of these models will be a key component of TAP field activities. In carrying out these tasks TAP will work closely with other programs in DOE and in other Federal and state agencies, and with the private sector. A forum to directly work with our counterparts in industry to ensure that the results of this research are translated into products that are useful to that community will be provided by NARSTO (formerly the North American Research Strategy on Tropospheric Ozone), a public/private partnership, whose membership spans government, the utilities, industry, and university researchers in Mexico, the United States, and Canada.

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1. Introduction

1.1 Tropospheric Aerosols and Their Importance

There is a compelling body of evidence that increasing concentrations of tropospheric aerosols from human activities are potentially major factors in human health and welfare.

Tropospheric aerosols are suspensions of fine particles, of diameter ranging from nanometers to micrometers, in the lower few kilometers of the atmosphere. These suspensions derive from "primary" sources involving direct emissions of particles, and by "secondary" processes, reactions of gaseous precursor emissions in the atmosphere to form particulate matter.

Fine airborne particles have been associated with adverse influence on human health in numerous studies. Much of the relevant research is summarized in the 1999 Draft EPA Criteria Document for Particulate Matter (EPA, 1999a), which identifies the following effects: (1) lung function decrements; (2) respiratory symptoms, or exacerbation of symptoms requiring bronchodilator therapy; (3) hospital admissions for respiratory and cardiovascular causes; (4) emergency medical visits; and (5) death largely from cardiopulmonary causes in the elderly. {pp 8-33 - 8-34]. The document presents a detailed examination of health effects associated with ambient particulate matter, examining the results of several studies that attempted to quantitatively determine the relative risk associated with a given concentration of PM. That examination concluded that "All of these long-term studies report many statistically significant findings associated with long-term mean PM concentrations. " [page 8-32]. However there remain important unresolved questions about the mechanism of insult, and whether the insult depends solely on the mass concentration of particles or on their chemical constituents.

Aerosol particles are also responsible for visibility impairment not just in urban areas and multi-city complexes, but also over widespread areas that encompass National Parks and wilderness areas for which it is desired to protect and enhance air quality related values. Aerosol particles and their precursors are the carriers of a number of hazardous air pollutants, and are the chemical agents responsible for acid deposition. They are also recognized to exert a major influence on the shortwave radiation energy budget of the Earth, and in the aggregate anthropogenic aerosols may be offsetting a major but unknown fraction of the anthropogenic greenhouse effect.
Of these several issues, the issue of aerosol influences on human health is viewed as having the highest priority in setting research directions and priorities.

Fine airborne particles are associated with respiratory disease, visibility reduction, acid deposition, and climate change. Of these issues, the issue of aerosol influences on human health assumes the highest priority for setting research directions.

Evidence suggesting adverse consequences for human health of airborne particles has led to the establishment of a series of National Ambient Air Quality Standards (NAAQS) designed to reduce the risks of breathing polluted air. The most recent NAAQS established in 1997 has objectives for clean air based on fine particle mass concentration. The controversy surrounding the scientific basis for this standard has resulted in Congressional direction to re-examine and elaborate on this scientific information. Atmospheric processes are central to this re-examination because of their critical role in determining aerosol properties governing their health effects.

In response to this direction, agencies of the government have initiated a national program to investigate the origins, evolution and health consequences of fine particles and their

chemical constituents. Much of the impetus for the direction of fine particle research lies with the Committee on Research Priorities for Airborne Particulate Matter of the National Research Council (NRC, 1998; 1999; 2001).

This enterprise is coordinated by the Federal Interagency Air Quality Research Subcommittee (AQRS), and much of the research is implemented in collaboration with NARSTO (formerly North American Research Strategy for Tropospheric Ozone, but now simply NARSTO, in recognition of its expanded effort directed to particulate matter; see NARSTO, 1999). NARSTO is a public/private partnership, whose membership spans government, the utilities, industry, and academe throughout Mexico, the United States, and Canada. Its primary mission is to coordinate and enhance policy-relevant scientific research and assessment of tropospheric pollution behavior, with the central programmatic goal of determining workable, efficient, and effective strategies for local and regional air-pollution management.

The evolving US national program focusing on the risks and management of the health effects of fine particles has four major components:

- **Health Science.** Establishment of a systematic approach to substantially advance knowledge about the mechanisms and specific insults leading to health consequences of exposure to fine particles in ambient air. ***The health science component recently has been established through the efforts of the Environmental Protection Agency (EPA) following the recommendations of the NRC Committee.***

- **Monitoring Networks.** Establishment of a hierarchy of measurement sites nationwide for monitoring the mass concentration, chemical composition and size distribution of fine particles. ***A national network of measurements and air monitoring has been designed and is being implemented through the EPA, the Department of Agriculture, the Department of Interior, and state environmental authorities.***

- **Emissions.** Establishment of a program to determine the emissions of fine particles and their gaseous precursors that will permit the preparation of much more effective management strategies. ***The investigations leading to improvement in knowledge of emissions involve work of EPA, the Department of Energy (DOE), state and local air quality management agencies, and the private sector, including the electric utilities (e.g., EPRI, the Electric Power Research Institute), the fossil fuels industry (e.g., American Petroleum Institute) and the transportation industry (e.g., Coordinating Research Council).***

- **Atmospheric Processes.** Improving knowledge about the atmospheric processes that govern the characteristics and evolution of airborne particles so that methods for quantitatively linking emission with exposure can be made available for management purposes. ***The investigation of tropospheric aerosol processes currently is supported by a number of sponsors in the public and private sectors, including DOE, the National Atmospheric and Oceanic Administration (NOAA) and the National Science Foundation (NSF).***

These four components of the national program are coordinated through the AQRS and NARSTO. A compilation of particulate matter research activities in the United States is maintained by the Health Effects Institute (HEI, 2001).

Critical Mass. Much progress has been made recently in initiating programs to meet the national needs through the first two components, health science and monitoring networks. Progress has also been made in the emissions arena, however accurate emission inventories remain a large area of uncertainty in linking what is present in the air to what is emitted by natural and anthropogenic sources. Similarly, while much pioneering research in atmospheric processes has been accomplished by NOAA and other agencies, including DOE, these efforts have not yet reached critical mass.

There are substantial and critical gaps in knowledge and understanding of the atmospheric processes associated with aerosols, and so the Tropospheric Aerosol Program (TAP) is being developed to complement on-going efforts at NOAA and other agencies, to help fill those gaps.

The research program presented here, the Tropospheric Aerosol Program (TAP), will be a sustained and cohesive means of investigating atmospheric processes affecting airborne particles, focusing on the atmospheric transformation processes that govern the mass concentration, chemical composition, and microphysical properties of fine atmospheric particles.

TAP will therefore specifically address the fourth research component of the national program, improving knowledge of atmospheric processes governing loading and properties of fine atmospheric particles. TAP will take advantage of new measurement technologies for gases and particles that have emerged in recent years and will enhance the state of the art of measurement and characterization of fine particles. TAP will thus contribute substantially to the measurement data base of loadings and properties of fine particles, thereby considerably augmenting the second component of the national fine particle research effort noted above.

Because the atmospheric loading of fine particles derives in great measure from energy production and use, and because of DOE's statutory responsibility to conduct research into the environmental consequences of energy-related activities, DOE is prepared to organize and act as the principal agent for TAP.

This Program Plan presents an overview of the research to be conducted in TAP. This plan is a natural outgrowth of reviews, assessments, and workshops undertaken over the past few years by the NRC (NRC, 1998, 1999, 2001), the AQRS (AQRS, 1998, 1999), NARSTO (NARSTO, 1998a, b, c; Hales, 1998; Hidy et al., 1998), and by several of the DOE laboratories (PNNL, 1999).

This plan was prepared at the initiative of the DOE Office of Biological and Environmental Research (OBER) (within the DOE Office of Science) in response to these crucial national and energy-related needs. These efforts were also encouraged by and coordinated with colleagues in the DOE Office of Energy Efficiency and the DOE Office of Fossil Energy.

After a draft version of this document had been circulated, a workshop was held at Brookhaven National Laboratory in June, 1999, to gain input from a broad community representing scientists from DOE National Laboratories, other Federal laboratories, academia, and the private sector, and officials in the various Federal agencies responsible for air quality and aerosol research. A list of participants is given in Appendix A.

TAP will focus on the atmospheric transformation processes governing aerosol loading, composition, and properties.

Following the Workshop a Preliminary Program Plan was prepared incorporating input from workshop participants and others to whom the draft had been made available. That document was circulated to Workshop participants and to other scientists and officials in the several cognizant Federal agencies and was made available on the World Wide Web. This Program Plan is a refinement of that document that takes into account comments and suggestions on the Preliminary Program Plan.

1.2 Priority Components of TAP

The principal objective of TAP is to determine the role of atmospheric processes in governing the physical and chemical characteristics of aerosols that are responsible for human health effects. These processes are the link between the emissions that are responsible for atmospheric aerosols and affected individuals who breathe them. Mathematical models of these processes are the primary tools scientists use to organize scientific knowledge into a comprehensive understanding of these links between emissions and exposure.

Highest priority components of TAP research are chemical composition of fine particles as a function of size, chemical dynamics and co-pollutant interactions, and estimating the response of fine particle properties to changes in particle and gaseous precursor emissions.

They are also the tools that regulators must use in determining how changes in emissions might affect human exposure to atmospheric aerosols and, consequently, their human health effects. Construction of models that relate emissions of air pollutants to ambient air concentrations and to their physical and chemical characteristics will, therefore, be a primary focus of TAP research.

Experience has shown that development and testing of these models is best achieved by conducting a series of field experiments, guided and supported by theoretical and laboratory studies, that focus on the key processes that govern aerosol formation, transformation, transport, and removal integrated by analysis and interpretation leading to preparation and evaluation of the models.

Table 1 lists critical elements of scientific knowledge that are needed to understand the links between emissions and exposure. Also included is an assessment, based on current knowledge, of the relative importance of these elements to human health concerns.

Recently representatives of different agencies of the Federal government and industry were polled to estimate the priorities (high, medium and low) that each of the components listed in Table 1 would have for advancing knowledge relevant to managing health risk from exposure to fine particles. The results are included in the table to illustrate the priority setting the TAP community proposes to refine in developing its design.

The results strongly suggest that the highest priority components for initial TAP planning

Table 1.1. Illustration of Institutional Priorities for Strategic Elements Addressing the Health Effects Associated with Airborne fine Particles

Critical element of scientific knowledge	Key to Health Concern	Uncertainty in Level of Current Knowledge	Link with Atmospheric Processes	Current contribution by Others	Proposed TAP Contribution
Number/surface/ mass-particle size distribution	H*	L (H for nanoparticles)	M	L	H
Chemical composition-size distribution	H*	M-H	H	M	H
Particle morphology (incl. internal-external mixtures)	H*	H*	L-M	L-M	M-H
Chemical dynamics and co-pollutant interaction	M-H*	H	H	M	H
Optical properties	L	M	M	M	M
Physical and meteorological processes	M-H*	H	H	M	H
Response of particle loading to change in emissions	H	H	H	M	H

Symbols H, M, and L denote high, medium, and low, respectively.

*Health research is expected to refine and improve requirements for knowledge in these areas before the final design of TAP.

include the chemical composition of fine particles as a function of their size, the chemical dynamics and co-pollutant interactions of fine particles, the physical and meteorological influences on fine particle concentrations, and the means to estimate the response of fine particle properties with changes in particle and gaseous precursor emissions.

In keeping with the present outlook of the AQRS, advancing the knowledge of regional haze and the component of aerosol forcing of climate change are designated second and third priorities for TAP. Because the origins of regional haze are closely linked with fine particle concentrations at the surface and aloft, new knowledge of the sources contributing to haze will emerge naturally from TAP experiments.

The field experiments envisioned in the TAP design will incorporate a major airborne sampling component. Thus important "targets of opportunity" will present themselves to conduct cooperative projects with atmospheric radiation research projects to determine the fine particle optical effects that bear on visibility and radiative transfer over urban and rural areas of the U.S.

1.3 Benefits to be Derived from TAP

The US has relied on a number of strategies to reduce particulate air pollution over the past twenty nine years. As air quality has continued to improve, decision makers have recognized that the management of major pollutants requires consideration of the consequences to one component with changes in another. The recognition of interactions between pollutants creates the need for highly complex conceptual models to minimize the risks of overly simplistic decisions for emissions reductions.

Although significant progress has been made in improving the nation's air quality, substantial costs have been incurred. The potential additional costs per increment of improvement in air quality are expected to rise significantly in meeting up-dated NAAQS. Therefore, it is critical to acquire new knowledge about the fine particles and their linkages to other air pollutants to insure that continued investment in air quality improvement will achieve the goal of greatly enhanced public health and welfare at minimal cost.

The research to be conducted by TAP, described in this Program Plan, together with that of other agencies, will provide major advances in knowledge about the processes that govern evolution of fine particles in the troposphere.

This knowledge will be used as a primary means to advance the development of greatly improved predictive capability for aerosol particle loading, properties, and geographical distribution that is required for informed decision making.

Decision making needs to rely increasingly on the ability to examine the consequences of alternative strategies to achieve a given air quality objective, which may take a form involving either mass concentration or chemical composition, or both. The principal means of achieving this objective lies in the application of reliable air quality models.

With the national program addressing in parallel the four major elements of the fine particle problem, and with TAP serving as a focal point for atmospheric processes, this will provide in the next decade a timely and complete portfolio for creating greatly improved air quality models relating sources and human exposure to fine particles, and co-pollutants. This knowledge will aid significantly in creating methods for selection of optimum strategies to reduce the health risks of exposure to fine particle in combination with gaseous components, including ozone, sulfur dioxide, and the nitrogen oxides.

The secondary benefits of TAP will include new knowledge about the optical properties of fine particles, as well as their evolutionary processes which, in turn, will fill gaps in important knowledge needed for considering

reductions in regional haze, and for particle influences on climate alteration.

1.4 Consequences of Not Doing this Research

The nation has committed major resources to resolve the human health consequences of exposure to fine particles in the atmosphere. This problem is an exceedingly complex one that has eluded effective solutions despite a long history of research and air quality management practice over the past thirty years.

The necessary resources have been committed through recent Congressional and administration actions to attack the health consequences of fine particle exposure. However, a comparable commitment for the parallel development of the necessary, advanced tools to inform decision makers about ambient air management is not yet available.

The capabilities of the scientific community are commensurate with the formidable task of obtaining this kind of information, but need to be focused in cohesively through a cooperative and collaborative initiative such as TAP, in the framework of the national program.

The timing of investigations leading to major improvements in mathematical models for quantifying emissions to ambient air concentration (and human exposure) is critical to insure that timely strategies can be developed as early as possible to minimize the risk of delaying pollution management decisions, or taking a less than optimal direction in management approaches.

If the national fine particle program engages in research on only two of the four major strategic elements, the necessary advanced information for decision making will be delayed far into the future. TAP offers the opportunity, at a level of $20-40 million a year, to focus the necessary scientific resources to address the fourth component of the national research program on fine particles.

At this critical level of commitment, initiating TAP at this time will insure that its products will be available for strategy development in the period 2005-2011. This is the period when the NAAQS for fine particles will be reviewed again in its five year cycle, and when critical, major decisions will be made for implementation of national efforts to reduce airborne particle exposures simultaneously with resolving parallel problems associated with tropospheric ozone, and regional haze.

Without the benefit of results from TAP there is a significant risk of choosing a non-optimum and hence less efficient or more costly pathway to a combined solution for these problems.

2. Overview and Background

2.1 Development of TAP

The Tropospheric Aerosol Program (TAP) is a program being developed by the Office of Biological and Environmental Research (OBER) of the Department of Energy directed to gaining improved scientific understanding and model-based representation of the processes controlling the mass loading, geographical distribution, chemical composition, microphysical, and optical properties of tropospheric aerosols. This program is a continuation of DOE's effort to develop understanding and predictive capability for the atmospheric influence of energy-related activities and their effect on human health and welfare. The TAP program is a highly focused observational and analytical research effort that will compare observations and model calculations to improve this modeling capability within known and reasonable accuracy.

This Preliminary Program Plan specifies the objectives of TAP and presents the need for this understanding and capability to represent these processes in atmospheric models. The Plan then presents the approach that will be taken by TAP to conduct the research necessary to develop this understanding and model-based representation.

2.2 Fine Atmospheric Particles

Aerosol particles in the atmosphere result either from introduction of particles into the atmosphere (primary aerosols) or from conversion of gaseous substances in the atmosphere to particulate matter (secondary aerosols). Aerosols result from natural processes and from human activities. Examples of natural aerosols are sea salt resulting from breaking waves, windblown mineral dust, smoke from wildfires, and the natural haze that is recognized in place names such as the Blue Ridge Mountains. Aerosols from human activities include smoke and diesel exhaust as well as visibility reducing "industrial haze" or "smog." In cities and nearby regions anthropogenic aerosols (that is, aerosols resulting from human activities) dominate aerosol loading.

Motivated by the desire to protect human health, the U.S. has recently adopted a new National Ambient Air Quality Standard (NAAQS) which for the first time sets a limit on the mass loading of particulate matter of aerodynamic diameter of less than 2.5 μm (termed the PM-2.5 or fine particle standard). The restriction of this standard to fine particles takes into account the understanding that fine particles are capable of penetrating well into the lungs with the resultant possibility of health impairment. Two standards have been issued: annual average concentration, 15 μg m^{-3}, and 24-hour average, 65 μg m^{-3}. The need to achieve the national standards is based on an increasing body of evidence that indicates that fine particulate matter in the atmosphere is responsible for adverse human health effects. The increased focus on the fine particles comes as a result of advancing knowledge of the apparent risk of inhaling pollutant aerosols with the potential for exacerbating respiratory ailments and other diseases.

The PM-2.5 standard, as is the case for ambient air quality standards generally, imposes a requirement on states and other jurisdictions to achieve a specified mass

TAP is a highly focused observational and analytical research effort that will compare observations and model calculations to improve capability of modeling aerosol loadings and properties within known and reasonable accuracy.

New Ambient Air Quality Standards for the first time limit mass loadings of fine particles.

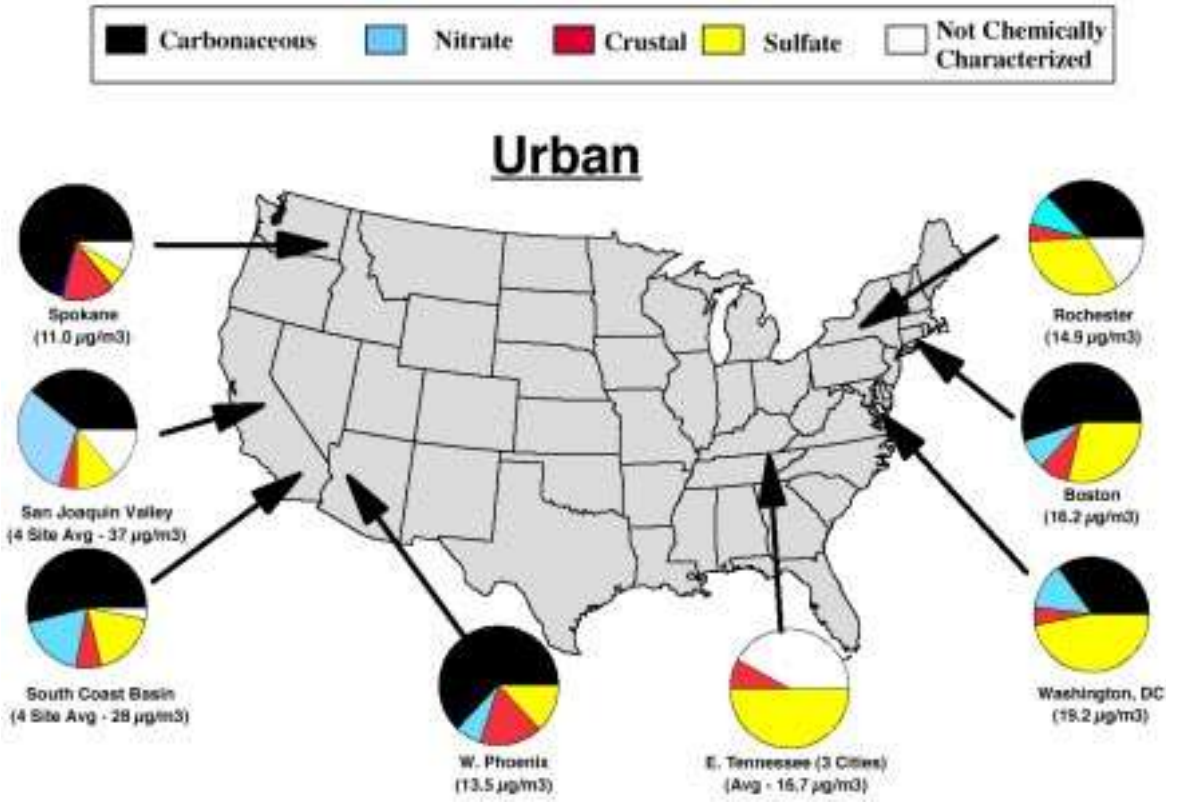

Figure 2.1. Annual average mass-loading and composition of sub 2.5-µm aerosol based on at least one year of monitoring at two or more sites in the same region. Data were obtained using a variety of non-federal reference methods and should not be used to determine compliance with the PM-2.5 NAAQS. From EPA (1998).

Because of differences in aerosol properties in different regions there can be no universally effective means of achieving the PM2.5 standard.

loading of aerosols in ambient air but does not specify a means of achieving this standard. Devising such a strategy requires at minimum an understanding of the source-receptor relation that is responsible for the aerosol loading at a given location. This in turn requires answering such questions as the relative contribution to aerosol loading of primary and secondary aerosols, and the relative contributions of natural and anthropogenic aerosols. Figure 2.1 shows large differences in the composition of sub 2.5 µm aerosol as a function of geographical location. It is necessary as well to understand the geographical dependence of aerosol properties and loading, as governed by rates of formation and removal, dilution, and transport in the atmosphere.

Present information on aerosol loadings in the United States indicates that exceedances of the PM-2.5 standard will be widespread, although the extent of such exceedances is not known in detail because there is not yet a widespread database of measurements performed according the EPA reference method for fine particles. The EPA has embarked on a major measurement program of fine particle mass loading in order to develop such a data base (EPA, 1999b). It is clear however that simply establishing the extent of exceedances is only a first step to developing a compliance plan. Achieving compliance with the new PM-2.5 standards will require understanding of the processes responsible for fine particle loading at a given location, the sources responsible for emissions of fine particles and their gaseous precursors and their rates and geographical distribution, the atmospheric transformation processes responsible for modifying the amount and nature of aerosol material, and the transport

and dispersion processes that govern local concentrations, in brief the processes responsible for aerosol loading at a given location. Figure 2.1 makes it clear that there will be no single answer to these questions that is applicable to the entire country. Likewise it is reasonably anticipated that the answers to these questions will depend also on season as a consequence of seasonal dependence of the mix of anthropogenic and natural emissions and also seasonal differences in controlling meteorology and atmospheric chemistry.

Understanding the processes that control the loading, distribution, and properties of submicrometer aerosols is going to be difficult. Aerosols are chemically and physically much more complex than gaseous pollutants, and the overall process governing aerosol loadings, properties, and distributions is much more complex than for primary pollutants such as SO_2 and CO. Aerosols are very heterogeneous in composition and sources, ranging from seasalt, dust and tire particles, to sulfates, nitrate, organics and soot, as well as mixtures of these materials. Some of the aerosols are emitted directly as particles, whereas others form in the atmosphere from gaseous precursors, both anthropogenic and natural. While in the atmosphere aerosol particles grow in size and evolve in composition through adsorption and reaction of gases and through coagulation and can change phase by deliquescence and efflorescence. They are removed through wet and dry deposition processes which are dependent on the composition and size of the particles.

There are numerous activities required to understand and quantitatively describe the loading and properties of sub 2.5 μm particles, from generating emission inventories to regional monitoring, to developing the knowledge of the fundamental processes that control loading of tropospheric aerosols and concentrations of specific classes of compounds. The latter processes, which constitute what might be denoted the "***atmospheric life cycle***" of these aerosols, are the principal focus of TAP. A recent NRC report called for development of advanced mathematical, modeling, and monitoring tools to represent the relationships between specific sources of particulate matter and human exposures and for linking sources of toxicologically important constituents and characteristics of particulate matter to exposed individuals and populations. (NRC, 1998). But this cannot be done without understanding of processes that control these relationships. This is where DOE comes in, with expertise in atmospheric aerosol science, in conducting large-scale field projects, in numerical modeling of atmospheric chemistry phenomena, and in laboratory studies and theory. In this effort we fully expect to complement and leverage other federal programs, principally those of EPA, NASA, NOAA, and NSF. These agencies have all declared PM-2.5 research as a high priority research focus for the near future. As has been shown in the past, coordinated work by multiple agencies is the key to real progress.

TAP will focus on the life cycle of atmospheric aerosols.

Closely related to the PM-2.5 issue is the issue of visibility impairment due to aerosols. As recently as April 22, 1999, Vice President Gore along with Environmental Protection Agency Administrator Browner announced a major new effort to improve air quality in national parks and wilderness areas. The new regional haze rule (Code of Federal Regulations, undated) requires states to develop implementation plans to prevent impairment of visibility in National Parks and other pristine locations, with the objective, ultimately, of attaining natural visibility conditions by 2064. The principal contribution to visibility impairment is from fine (sub 2.5 μm diameter) particles, which are highly efficient scatterers of visible radiation and whose concentrations are closely correlated with the atmospheric light scattering coefficient (Figure 2.2). Whatever the ultimate regulations that are imposed to protect and improve visibility, it is clear that developing a strategy to achieve any such standards requires the same understanding of the processes that control tropospheric aerosol loading and properties as is required to develop strategies to meet the PM-2.5 standards.

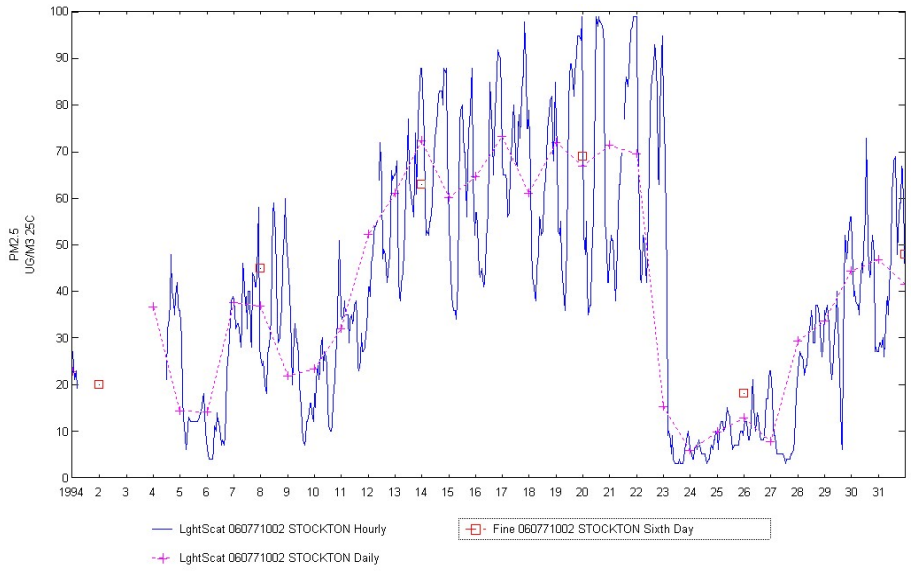

Figure 2.2. Time series of light scattering coefficient and fine particle mass in Stockton CA in January 1994. Light scattering coefficient is scaled to fine particle mass by daily averages. From Husar (1997).

Another related environmental issue is acid deposition. The recent report from the National Acid Precipitation Assessment Program (NAPAP, 1998) indicates that it is too early to determine whether changes in aquatic ecosystems have resulted from emission reductions in response to the 1990 Clean Air Act Amendments. The report notes that over the last 13 years New England lakes have shown evidence of recovery from acidification but in contrast that the majority of Adirondack lakes have remained fairly constant and that the most sensitive Adirondack lakes have continued to acidify. The EPA has reported to Congress that additional reductions in sulfur and nitrogen deposition will be required to fully restore sensitive Adirondack lakes. Sulfate and nitrate, major components of fine particle mass (Figure 2.1) and their gaseous precursors sulfur dioxide and nitrogen oxides are the major contributors to acid deposition. Figure 2.3 shows that concentrations of aerosol sulfate and of nitric acid vapor, a major aerosol precursor, show no indication of decrease over the past 12 years in nonurban regions of the Northeast.

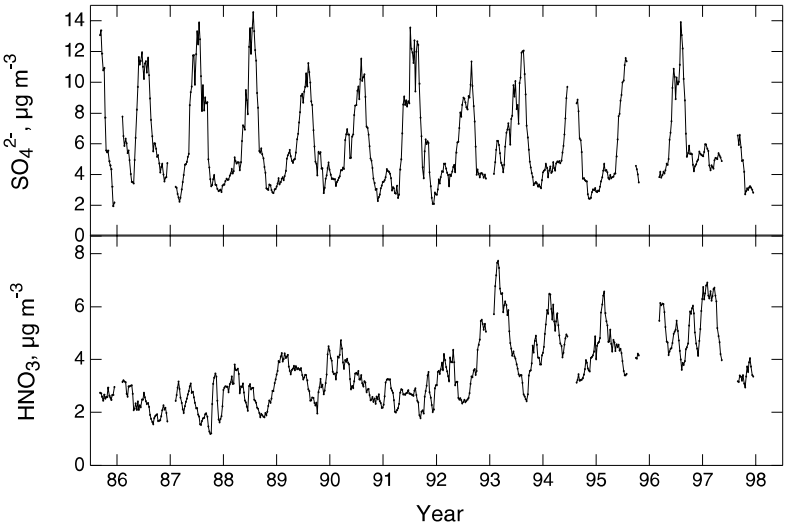

Figure 2.3. Time series of concentrations of sulfate aerosol and nitric acid vapor at a site near State College, Pa. Adapted from NAPAP (1998), with update from B. Hicks, NOAA, Air Resources Laboratory (private communication, June 1999). Data points are 5-week running mean of 1-week samples.

Lastly, research in the last decade has indicated that aerosol particles have a potentially significant influence on radiative energy exchange processes in the atmosphere. Anthropogenic aerosols scatter solar radiation and modify cloud reflectivity, thereby exerting a cooling influence on climate. This cooling influence is thought to be offsetting a substantial fraction of anthropogenic greenhouse warming, but its magnitude is highly uncertain. This uncertainty precludes confident empirical detection of climate change due to increased greenhouse gases and quantitative inference of climate sensitivity. It also precludes evaluation of performance of global climate model simulations so that it is impossible at present to answer the question of how much of the 0.6 degree temperature rise over the past century can be ascribed to industrial greenhouse gases and how much to natural variation. Much of this uncertainty arises from present limited ability to quantitatively describe the aerosol loading and microphysical properties that govern their light scattering and cloud nucleating ability.

Shortwave radiative forcing of climate change by aerosols by the direct and indirect mechanisms has been identified by the Intergovernmental Panel on Climate Change (IPCC) as the greatest uncertainty in forcing of climate change over the industrial period (IPCC, 1996). In 1996 a NRC panel, convened at the behest of DOE, NASA, NOAA, and NSF, issued an urgent call for a coordinated program of research to quantify this aerosol forcing (NRC, 1996). Since that time rather little has been initiated. As we get ready to face the next millennium and draft a policy to mitigate or accommodate to the anticipated climate change, it is imperative to resolve this largest scientific uncertainty in climate forcing.

While research programs that deal with climate change, including the DOE Atmospheric Radiation Measurement Program (ARM, 1999; Stokes and Schwartz, 1994), examine the radiative effects of aerosols, there is a major gap in research to quantitatively describe the life cycle of tropospheric aerosols as it pertains to this radiative forcing, research that is more properly identified as in the domain of atmospheric chemistry and which builds on the techniques and capabilities of this discipline. Understanding the radiative influence of aerosols requires field studies that provide a seamless link of understanding connecting aerosol emissions, secondary formation and evolution, chemical, physical, and optical properties, and deposition together with examination of radiative influences. Ultimately what is required are models that accurately represent aerosol loading, i.e., the amount and geographical distribution of aerosols in the atmosphere, and the so-called intensive properties of these aerosols, the microphysical and optical properties that govern their radiative forcing per mass loading. At present representation of aerosol processes in climate modeling is in its infancy, and to the extent that this is done at all, it is done in a quite simplistic way that does not take into account the heterogeneity of size and composition that is characteristic of aerosols. For example because of lack of knowledge and model based representation of aerosol composition and properties, there has thus far been a tendency, to the extent that aerosol properties are addressed in models at all, to treat all aerosols as if they were composed of ammonium sulfate at 25°C. However it is known from recent results that aerosol deliquescence and hygroscopic growth, and hence optical properties, depend markedly on composition and temperature. It is therefore necessary to develop the knowledge base that can handle the complexity of actual aerosols.

Uncertainties due to aerosols preclude quantitative attribution of climate change over the past century to industrial greenhouse gases.

There is a major gap in research to describe the life cycle of tropospheric aerosols as it pertains to radiative forcing of climate change.

It seems likely that solving the greenhouse-gas climate change issue will also require a significant change in the nation's energy economy at a cost of billions of dollars. Moreover, by virtue of the fact that aerosols couple air quality with climate change our actions to overcome one of these challenges will affect the other. In this context it is urgently imperative that the aerosol forcing be placed on a much more quantitative foundation than is available at present.

The management of these interrelated environmental stresses poses a major challenge to decision-makers, in terms of both

the atmospheric processes and the mechanisms of effects on humans and the environment. Recently the United States EPA has created a major goal to document the nature of ground-level airborne particles and has challenged the health and atmospheric science community to identify the specific atmospheric agents responsible for adverse effects of particulate pollution [EPA, 1999b] These initiatives are superimposed on continuing research on visibility reducing pollution and on the influence of particles on climate alteration. The Tropospheric Aerosol Program (TAP) will complement the initiatives just under way in the monitoring and health effects areas, and add substantially to the capabilities for advancing methods for predicting the response of atmospheric pollution exposure to emissions management options.

TAP must be viewed as one component of an integrated national program of research on atmospheric aerosols.

2.3 The DOE Context

The fact that much of the tropospheric aerosol burden is believed to result from energy production and use focuses attention on this key component of the nation's well-being in the context of meeting the nation's energy requirements in the next century. Understanding the environmental influences of energy production and use and ways to minimize these influences is a necessary component of DOE's National Energy Strategy, which has an explicit goal to promote energy production and use in ways that respect health and environmental values —improving our health and local, regional, and global environmental quality (DOE, 1998a).

Referring explicitly to PM2.5 particles former Secretary of Energy Bill Richardson has stated, "These unseen particles may pose respiratory problems for certain portions of the population, and for this Administration, there is no higher priority than protecting the health of our citizens. . . At the same time, if our clean air regulations are to be fair and scientifically-sound, we need to understand much better the linkage between the levels of these pollutants in the atmosphere and their sources, both human and natural." (DOE, 1998b). The TAP program is specifically directed to developing this understanding.

The objective of TAP should be viewed in the context of the national portfolio of measurements and research on atmospheric aerosols. There are many elements that must be implemented in order to be able to meet this country's requirements, and the several Federal agencies have differing responsibilities in this respect, although to be sure the boundaries that separate these responsibilities are somewhat ill defined. EPA's responsibilities include development and evaluation of emission inventories of aerosols and aerosol precursors, monitoring to determine the extent of compliance/non-compliance with air quality standards, and epidemiological studies to quantify aerosol health effects. NOAA has responsibility for long term monitoring of aerosol loading and properties at a small number of sites. Some of these elements are in place now and some of them are being enhanced. A national monitoring network will undoubtedly produce invaluable data, but without the supporting mechanistic knowledge, its value is very limited in providing guidance for developing an efficient control strategy or in evaluating present, past, or future aerosol influences on climate.

If the goal of a national aerosol program includes, as it should, developing the capability to devise effective strategies for control of aerosol loadings and the ability to quantitatively estimate aerosol influences on climate, it is essential to acquire fundamental understanding of the atmospheric transformation processes governing the loading and properties of tropospheric aerosols. This process-level understanding is fundamental to constructing models that are generally applicable, not just to a limited range of conditions, so that they may be applied to a wide variety of situations, for example modeling for previous emission scenarios in order to develop a historical record of forcing, or modeling for various emission scenarios to answer "what if" type questions. The DOE Atmospheric Chemistry Program (ACP, 1999) has demonstrated outstanding capability to perform process-level atmospheric research. To date this program has focused largely on the fundamental processes that control photochemical oxidants. TAP is viewed as quite complementary to the DOE Atmospheric

Chemistry Program and other DOE and federal research programs.

TAP must be viewed as one component of an integrated national program of research on atmospheric aerosols. It is essential that the programs of the several agencies be closely coordinated. The DOE component of the national program is process research to determine, quantitatively describe, and represent in models the mechanisms governing the mass loading, composition, and microphysical and optical properties of tropospheric aerosols, and their geographical and vertical distribution.

TAP will contribute to the DOE/OBER research effort directed to understanding the basic chemical and physical processes of the Earth's atmosphere and how these processes may be affected by energy production and use. This understanding will ultimately permit effective mitigation of the long-term health and environmental consequences of energy production and contribute to optimal use of differing technologies.

2.4 The Science Context for TAP

Although recognized for some time, the importance of tropospheric aerosols has recently been highlighted with respect to their influence on human health (NRC, 1998) and radiative forcing of climate change (NRC, 1996).

At present, despite a history of continued atmospheric research, there is insufficient information about the processes governing the composition and microphysical properties of tropospheric aerosols available to develop in a modern context insightful decisions to minimize the risk from these aerosols. The lack of information centers in the details of processes shaping the chemical composition as a function of particle size, especially regarding the carbonaceous and reactive nitrogen components, both of which are major constituents of fine particulate matter in numerous locations (Figure 2.1).

Examination of composition of tropospheric aerosol particles as a function of particle size indicates a wide range of variation, with much of present-day energy related contributions residing in the fine particle range. Combustion processes tend to produce very finely divided particles, formed either directly in the source or in the air near the source. These nanoparticles are present in large numbers near sources but are rapidly agglomerated in the air.

Health effects research over the years has indicated the importance of particle size in respiratory disease. The fine particle fraction of the particle-composition distribution has the greatest likelihood of affecting the lower lungs, where oxygen exchange takes place. Since recent health studies have identified sub-2.5 μm particles as a potential respiratory inflammatory agent, there is a need to characterize this fraction and its growth as well as the particle properties quantitatively for complex human exposure assessments. This kind of research is highly demanding of measurement capability since it requires scanning the particle size spectrum, which varies over at least three powers of ten in size, and more than seven powers of ten in concentration.

At present there is insufficient information about the processes governing the composition and microphysical properties of tropospheric aerosols to develop insightful decisions to minimize risk.

Other hypotheses advanced by the health effects research community relate observed health impacts to specific chemical factors including: metals, acids, organic compounds, biogenic particles, sulfate and nitrate salts, peroxides, soot, and gaseous cofactors (Albritton and Greenbaum, 1998). In order to make progress in establishing which of these agents are causative and the effects of PM control measures, it is necessary to understand the relations between emissions and the size-dependent composition of the ambient aerosol. Thus, demands of chemical speciation must be added to the task of determining size spectra over many orders of magnitude. Both measurements and predictive models are required.

The reactive nitrogen and carbonaceous components are closely linked with sulfur species and are interactive with oxidant production in the troposphere. Through the work of the last two decades the chemistry

governing gas-to-particle conversion of sulfur species is relatively well understood. The corresponding processes regarding particulate nitrogen and carbon are much less well understood and need to be brought to a similar level of understanding.

Quantitative description of the reactive nitrogen (ammonia-nitrogen oxide) gas-particle complex is incomplete. It is critical to understand the response of the sulfur-nitrogen-carbon system as changes in fossil fuel blends and their combustion emissions are mandated over the next few years. Current knowledge cannot specify quantitatively where all of the reactive nitrogen resides after emission into the air. There are apparent major differences in the nitrate content of aerosol particles in the eastern and western parts of the US. These are hypothesized to be related to conditions in which chemical equilibrium between nitric acid and ammonia exists in the presence of moisture, sulfate salts and carbonaceous material. However, this conjecture is based mainly on experience in southern California, and is generally unverified for conditions east of the Mississippi River. A second and equally important aspect from the health effects outlook is the potential for organo-nitrate compounds to be present in fine particles. The chemical processes in photochemical smog are known to produce gas-phase nitrate compounds that are potential eye and skin irritants. It is suspected but not established that organic nitrates are also present in particles in sufficient concentrations to be a factor in respiratory stress, possibly being a factor in more serious ailments.

The origins and composition of carbonaceous aerosols are poorly understood, despite their importance as a fraction of aerosol mass.

The origins and composition of carbonaceous aerosols are poorly understood, despite the importance of carbonaceous material as a fraction of aerosol mass. Evidence suggests that carbonaceous material, as either black (elemental) carbon or as organic material, makes up as much as half of the fine particle mass concentration in air sampled in the East and parts of the West (Figure 2.1). This component of aerosols has been difficult to measure precisely because it involves artifacts of sampling as well ambiguities of continuing exchange of semi-volatile compounds from particles suspended in the air.

Carbonaceous material in particles derives directly from combustion of fossil- and wood-based fuels. It also comes from the oxidation and subsequent condensation or adsorption of reaction products of volatile organic compounds. The volatiles come not only from anthropogenic emissions but also from exhalations from vegetation. Aside from the complexity of composition, two major unknowns exist. The first concerns how much production of organic material takes place from reactions of gaseous organic compounds; the second concerns how much of the secondary organic fraction derives from biogenic sources. Some have hypothesized that a substantial but unknown fraction of the haze that persists over the East in summer is made of secondary organics, which actively absorb moisture, as do most inorganic salts, such as ammonium sulfate and nitrate. Quantifying interactions of moisture with the complex mixture of organic species intermingled with inorganic chemicals is a major challenge for the scientists who hope to create reliable mathematical models for synthesizing scientific elements, and for air quality management.

Aerosol particles are known to be intimately involved in cloud droplet and ice particle formation and growth. At the same time cloud and precipitation processes are instrumental in changing the aerosol size distribution and in removing particles from the air. The micro-physical shaping of the particle-composition size distribution is a complex phenomenon which varies with cloud macro-scale processes. The sensitivity of cloud processes to aerosols has long been a subject of interest to atmospheric scientists. The quantitative coupling of the microscale phenomena to macroscale cloud and precipitation processes remains an unresolved problem, and the subsequent dilution or removal of particle populations, or the change in cloud optical properties is of great current interest for improvements in air quality models, and climate alteration calculations.

Even though progress has been substantial towards improving mathematical methods for estimating the response of the atmospheric processes to changes in man's energy practices, substantially more progress is

required. Progress will depend on a combination of major advances in knowledge through field experimentation, laboratory investigations, and synthesis of information into mathematical models.

2.5 The TAP Approach

Experience in atmospheric chemistry has shown that measurements cannot be sufficiently dense in space and time to meet all requirements, nor can measurements by themselves lead to predictive capacity necessary for developing approaches to meet air quality requirements. The situation is even more complicated in the case of atmospheric aerosols, which are highly heterogeneous from location to location (Figure 2.1) necessitating process-level understanding and precluding the utility of any "one size fits all" aerosol model. Likewise, empirical models will not be transferable to changing mix of emitted materials, changing atmospheric chemical environment or the like.

For these reasons TAP will consist of a sustained analysis and interpretation of results supported by a portfolio of tightly coupled research activities consisting of four principal components:

- Field Measurements of aerosol properties and transformation processes.
- Modeling transport and transformation of tropospheric aerosols.
- Development of instruments and advanced methods of aerosol characterization.
- Laboratory experiments and theory directed to aerosol processes.

The coupling of these several elements is indicated schematically in Figure 2.4. The field program is the centerpiece of TAP. The other elements are coupled not just to the field program but also to each other to maximize the synergism and utility of the entire TAP Program. These elements will enable the TAP scientific team to bring the atmospheric science of fine particles to the level necessary to bring informed energy and environmental management for the next several decades.

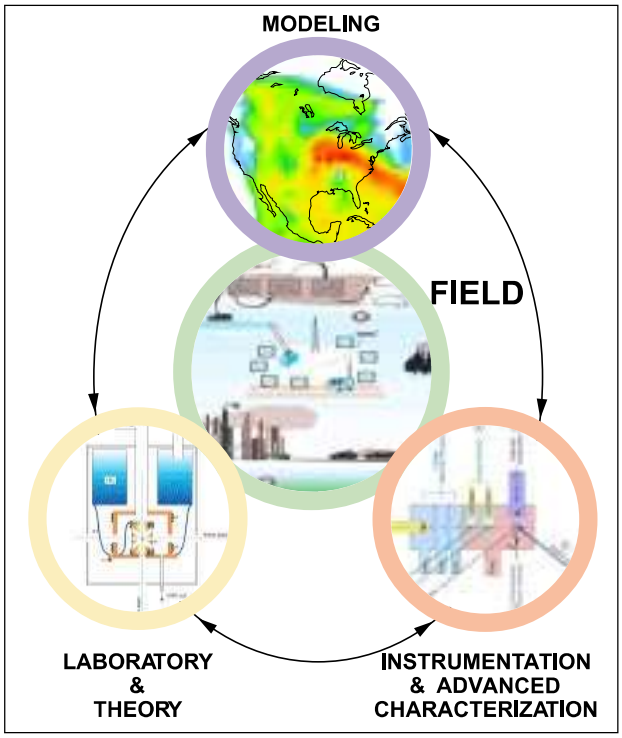

Figure 2.4. Science Elements of TAP.

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3. The TAP Objectives

3.1 What TAP will Accomplish

The goal of TAP will be to develop the fundamental scientific understanding required to construct tools for simulating the life cycle of tropospheric aerosols--the processes controlling their mass loading, composition, and microphysical and optical properties--all as a function of time, location, altitude, and ambient conditions. The TAP approach to achieving this goal will be by conducting closely linked field, modeling, laboratory, and theoretical studies focused on the processes controlling formation, size, chemical composition, optical properties, transport, and deposition of tropospheric aerosols. This understanding will be represented in models suitable for describing these processes on a variety of geographical scales, from tens to thousands of kilometers; developing and evaluating these models will be a key contribution of TAP. In carrying out these tasks TAP will work closely with other programs in DOE and in other Federal and state agencies, and in the private sector, directed to related aerosol issues.

3.2 TAP Research Tasks and Scientific Issues

The following are examples of the principal research tasks and scientific issues regarding tropospheric aerosols which will be accomplished by TAP or which the research conducted in TAP will resolve and/or provide the physical understanding and model-based representation to permit resolution in particular situations of interest.

- Characterize the size-dependent composition, microphysical properties, and other relevant properties of atmospheric aerosols, including optical properties, at the surface and throughout the vertical column, during TAP field projects.

- Determine the accuracy with which aerosol optical properties (light scattering coefficient, absorption coefficient, light-scattering phase function) can be calculated from knowledge of size distribution, chemical composition, and composition-dependent properties such as refractive index.

  Determine the accuracy of emission inventories for primary aerosols and for aerosol precursor gases in locations of TAP field projects.

- Determine the fundamental processes that control new particle formation in the atmosphere and the dependence of the rates of these processes on controlling variables.

- Determine the processes contributing to the accumulation of mass on pre-existing atmospheric aerosol particles and the dependence of the rates of these processes on controlling variables.

- Develop the ability to describe the interaction of atmospheric aerosol particles with water vapor and the dependence of this interaction on particle composition and surface properties and on ambient conditions.

- Determine the rate of dry deposition as a removal process for aerosol mass and number and its dependence on particle size and composition.

- Determine the influence of meteorological processes, such as turbulent mixing in convective systems and accumulation in high- and low-pressure systems, in-cloud reactions, precipitation scavenging, on

TAP will develop the fundamental scientific understanding required to construct tools for simulating the life cycle of tropospheric aerosols--the processes controlling their mass loading, composition, and microphysical properties--all as a function of time, location, altitude, and ambient conditions.

temporal variation of aerosol loading and properties.

- Determine the influence of aerosol loading and properties on gas phase chemistry as sites of heterogeneous catalytic reactions, as sinks of gas-phase species, and by changing the actinic flux.

- Develop model-based tools to determine the fraction of the aerosol mass observed at a given location that derives from primary emissions versus gas to particle conversion in the atmosphere.

- Develop model-based tools to determine the fraction of aerosol mass at a given location that is anthropogenic versus natural.

- Determine the geographical and vertical scale that is required for physical simulation models to describe PM-2.5 exceedances.

- Determine the spatial and temporal resolution required in models to represent aerosol loadings and properties on a regional scale in order to provide realistic estimations of effects of emission control.

- Determine the computing power required to represent aerosol loading, and size resolved chemical, and physical properties in simulation models.

- Develop methods to parameterize aerosol processes in regional to continental scale air-quality models and in global scale climate models.

- Determine the accuracy with which physical simulation models describe aerosol loading, composition, size distribution and optical properties.

4. Organizational Structure

4.1 Organizational Elements of TAP

The organizational structure of TAP is designed to meet the needs of a complex scientific research program that will consist of individual research projects all directed to a common set of goals, and to being responsive to the management requirements of DOE.

The management and organizational structure for the program as presently envisioned is sketched in Figure 4.1. The major features of the program's organization are as follows.

1. Direct management of the Program by a Program Director in the Environmental Sciences Division of DOE's Office of Biological and Environmental Research.

2. Interagency Coordination, primarily through the Federal Air Quality Research Subcommittee (AQRS) and the various NARSTO working groups, to ensure close coordination with other programs.

3. A Lead Scientist, who must be an aerosol scientist with broad experience, will have overall responsibility for the implementation of TAP, with consultation with DOE and with the Scientific Steering Committee, for the scientific leadership of the program and for representing the program within the scientific community.

4. The Science Team will consist of the Principal Investigators of the scientific projects comprising TAP. Science projects will be selected competitively through proposals in response to DOE Program Announcements, or through joint announcements with other agencies.

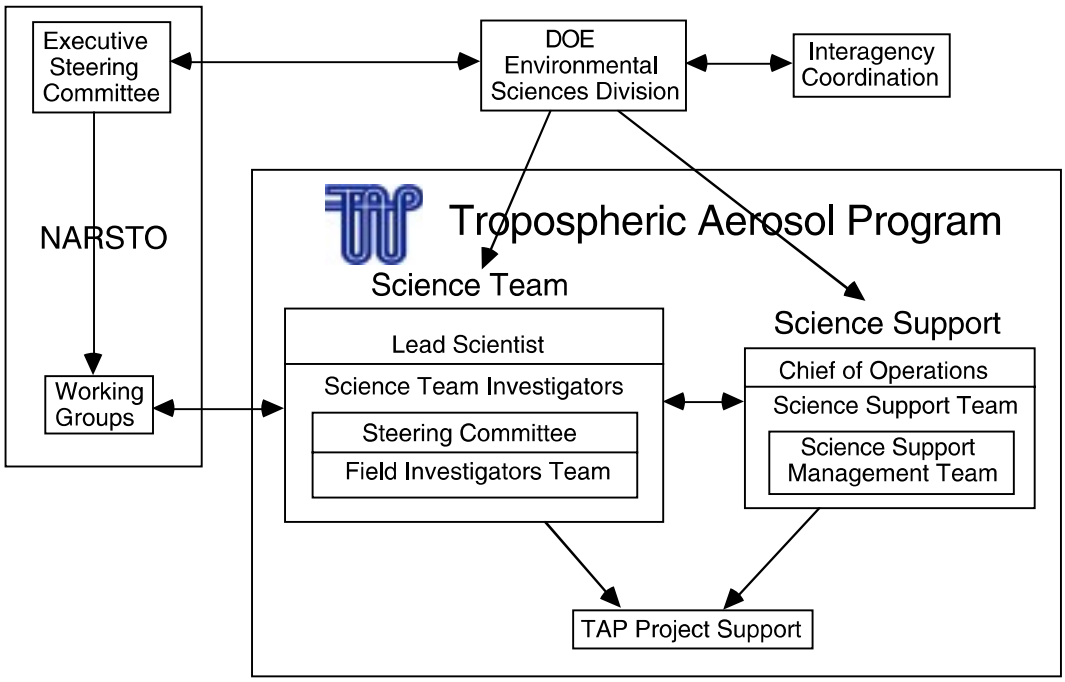


Figure 4.1. Organizational structure of TAP including DOE management oversight and Interagency Coordination, and showing linkage to the national research effort on tropospheric aerosols through NARSTO.

5. The Scientific Steering Committee will consist of representatives from each of the four programmatic areas of TAP and will, in conjunction with the Lead Scientist and the responsible DOE officials, set the scientific direction for TAP. The SSC will conduct periodic reviews of the set of TAP projects to ensure that key gaps in knowledge are being filled and to identify any research needs that are not being met. This will keep TAP focused on research critical to understanding PM2.5 and related issues for which where understanding of aerosol properties and evolution is required. The SSC will also function as a recruiting entity to insure that outstanding investigators participate in TAP and commit to getting "answers" that are needed.

6. The Field Investigators Team will consist of the principal investigators of field study teams, the principal scientific groups participating in the Field Campaigns of TAP, as described in Section 7. This team will define major field projects of TAP including selection from their membership of Project Scientists responsible for leading individual projects.

7. A Science Support Team under the direction of a Chief of Operations will provide support to TAP field and modeling operations. Science support resources will be allocated based upon requirements of the Science Team Projects with guidance from the Lead Scientist, the Steering Committee, and the Field Investigators Team, with concurrence of responsible DOE officials. The Science Support Management Team will consist of the heads of the several functional activities within Science Support.

8. TAP Project Support will be responsible for contracting and other fiscal matters and for public relations and outreach activities.

4.2 Science Team

The Science Team will consist of the project scientists selected based on peer review proposals to conduct specific projects as specified in DOE Research Announcements or through joint announcements with other agencies. Successful projects will be directed to answering key questions and to folding results into the requisite enhanced process-level understanding and ultimately into model-based representation of these processes. Care will taken in construction of requests for proposals and in award of grants that necessary research components are being addressed, without overemphasis of some components with underemphasis of others.
Science team members will be encouraged to participate in the various NARSTO and other-agency aerosol-related working groups, and scientists funded through other aerosol research programs will be invited to participate in TAP meetings and field campaigns.

4.3 Science Support Team

The Science Support Team is seen as crucial to achieving the objectives of TAP. This activity will have responsibility for:

Facilities. Provide and maintain aircraft, relocatable aerosol measurement facilities, instrumentation for field measurements.

Support for campaigns. Campaign planning and logistics; calibrations, measurement audits.

Support for modelers. Preparation of meteorological drivers and gridded data sets for comparisons; establishing and maintaining community models supporting swap in/out of modules.

Data system and archive. Incorporate measurements into data base/archive to facilitate model development and testing.

The requirements and activities of the several science support components are presented in Chapter 5.

4.4 Integration

An underlying philosophical tenet of TAP is that good science must undergo the scrutiny of peer review, in the selection of projects and in the publication of the findings of research. Traditionally in science the latter is through peer-reviewed journals, and this will certainly be a requirement for TAP investigators. It may reasonably be anticipated that over the duration of this program such publication will increasingly be by means of electronic publications, which are already being implemented in the geophysical sciences, and which maintain the rigorous requirements of peer review while at the same time permit publication of much richer data sets and even the dynamics of models, in contrast to the static format of traditional paper journals. TAP will actively encourage publication of findings in such media. TAP will also provide a web-based server to facilitate dissemination of publications reporting TAP research. This web site will serve as an active vehicle for communication among TAP investigators, for planning of field programs, for exchange of data and models, for preparation of reports, and for active exchange of ideas that will make TAP a truly collaborative program.

As a mission-oriented program, TAP has a responsibility to DOE to make its findings available to the user community in a way that will maximize the utility and use of these findings. A specific example is models. These should be made available in readily accessible electronic format that will allow their use by the external scientific and air quality management community. Likewise, many of the field measurement data of TAP will be of broad interest not just to TAP investigators but also to a variety of outside entities. To this end TAP will facilitate dissemination of research results by suitable web-based approaches.

TAP will conform to the data management policy of the U.S. Global Change Research Program (USGCRP, 1991). This policy calls for continuing commitment to the establishment, maintenance, validation, description, accessibility, and distribution of high-quality, long-term data sets, full and open sharing of data, preservation of data, inclusion in data archives of information about the data holdings, including quality assessments, supporting ancillary information, and guidance and aids for locating and obtaining the data, adherence to data dissemination standards, and timely availability of data subject to a reasonable period of exclusive use by program investigators.

4.5 Oversight and Interagency Coordination

Overall management responsibility for the Tropospheric Aerosol Program (TAP) is provided by the DOE Program Director.

Oversight is provided by DOE's Environmental Sciences Division (ESD), Office of Biological and Environmental Research (OBER), and the Biological and Environmental Research Advisory Committee (BERAC).

Interagency coordination is provided through the Federal Air Quality Research Subcommittee (AQRS), the Atmospheric Chemistry Panel of the Federal Subcommittee on Global Change Research (SGCR), and an interagency Aerosol Working Group.

4.6 Relation to Other DOE Programs

TAP will be closely linked to ongoing DOE projects dealing with the environmental influence of energy activities. The Atmospheric Chemistry Program deals with regional to global scale global chemistry and fate of tropospheric air pollutants. The Environmental Meteorology Program deals with measurement and modeling of vertical transport and mixing processes in the lowest few kilometers of the atmosphere. The Atmospheric Radiation Measurement Program is aimed at improving the understanding of the transfer of solar and terrestrial infrared radiation in the atmosphere, and the

TAP will be closely linked to ongoing DOE projects dealing with the environmental influence of energy activities. TAP complements these programs by focusing on the processes that control the loading, geographical distribution, chemical composition and microphysical properties of tropospheric aerosols, especially anthropogenic aerosols.

atmospheric properties controlling this radiation transfer and the representation of radiation transfer in climate models in the context of the necessity to represent climate change due to anthropogenic greenhouse gases and aerosols.

The TAP program complements these programs by focusing on the processes that control the loading, geographical distribution, chemical composition and microphysical properties of tropospheric aerosols, especially anthropogenic aerosols. Specifically, TAP focuses on atmospheric transformation processes governing the composition of tropospheric aerosols especially gas-to-particle conversion in the atmosphere, nucleation of new particles, growth of existing particles, and how these processes affect aerosol composition and properties.

Atmospheric Chemistry Program (ACP). The ACP consists of a set of research projects focusing on chemistry and fate of tropospheric air pollutants including aerosols on regional to global scales (ACP, 1999). Much ACP field work is directed to atmospheric oxidants and related free radicals. Because of the strong coupling of oxidant and aerosol chemistry, it is expected that some field projects in TAP may be conducted in conjunction with ACP field projects to take advantage of chemical and meteorological measurements that are pertinent to the requirements of both projects.

Atmospheric Radiation Measurement (ARM) Program. The Atmospheric Radiation Measurement (ARM) Program is directed to measurement and model based representation of radiative transfer in the earth's atmosphere and to the processes controlling radiation transfer, principally involving water vapor and clouds, and to lesser extent aerosols (ARM, 1999; Stokes and Schwartz, 1994). The ARM program maintains a highly instrumented site in north central Oklahoma at which a variety of measurements are employed to provide detailed characterization of the atmospheric state and meteorological variables that control the evolution of the atmospheric state. continuous measurements also include aerosol optical depth (during the daylight hours when the sun is visible, vertical distribution of aerosol, by Lidar, and aerosol optical properties and limited chemical variables at the surface. Because of these measurements it may be quite attractive to conduct one or more TAP field studies at the ARM Oklahoma site.

Environmental Meteorology Program. The measurement and modeling of vertical transport and mixing processes in the lowest few kilometers of the atmosphere are problems of fundamental importance and of much practical importance governing the accumulation of air pollutants, for which a fully satisfactory treatment has yet to be achieved. In recognition of this DOE ESD has recently initiated a Vertical Transport and Mixing Program (VTMX, 1998) directed to investigation of vertical transport and mixing processes in the lower atmosphere, concentrating on processes in stably stratified conditions, in conditions of weak or intermittent turbulence, and during morning and evening periods that mark transitions between stable and convective conditions with particular interest in urban regions affected by adjacent elevated terrain (e.g., urban basins or valleys). Because of the necessity for detailed characterization of the meteorological situation at the times of TAP measurements it is particularly attractive to consider conducting TAP field campaigns in conjunction with VTMX field campaigns.

DOE Research Aircraft Facility. Measurements aloft are critical to the success of TAP. A Gulfstream 159 (G-1) aircraft (DOE Research Aircraft Facility, 1998), outfitted with instrumentation for trace gas, aerosol, and meteorological measurements, can be deployed to field study locations. With a range of ~1,600 km and duration of ~4.5 hours, it can provide spatial distributions of ambient variables at altitudes from ~300 m to ~7.5 km across the regional scale of interest to TAP. The facility can also be used to flight test new instrumentation systems developed within TAP.

DOE Environmental Molecular Sciences Laboratory (EMSL). The EMSL is a national scientific user facility whose mission is to provide advanced and unique resources to scientists engaged in research on critical

environmental problems. It includes capabilities that are particularly well suited for research on the detection of aerosol precursors, and on formation, growth, reactivity, and surface and bulk species characterization for natural and model aerosols. It also provides massively parallel computational capabilities well suited to atmospheric sciences research. These facilities (EMSL, 1999) are available without charge for nonproprietary research.

DOE National Energy Technology Laboratory (NETL) Upper Ohio River Valley Study. In this study (DOE, 1998b) four monitoring sites in the region are being equipped with a broad array of sophisticated equipment, both commercially-available and still-experimental monitoring devices, to collect and analyze the small particles. Two of the locations will be so-called "supersites" and will be outfitted with devices to measure the chemical make-up, size and seasonal variations of the airborne particles. At each "supersite," meteorological data, such as wind speed and direction, relative humidity, and ultra-violet radiation, also will be gathered.

DOE NETL "Fingerprint" study. This study (DOE, 1998c) will examine whether fine particulate emissions from coal-burning systems have unique "fingerprints" that can identify their source. Researchers will trace the physical and chemical properties of PM2.5 to the properties of the coal burned, the operating conditions of the combustor and boiler, and the configuration and operation of pollution control technologies.

DOE Office of Heavy Vehicle Technologies' Diesel Particulate Sampling Methodology project. This project is motivated by the new PM-2.5 NAAQS and alternative fuels. The project involves determining actual particle size distribution and particle number concentrations in the exhaust plumes from heavy-duty diesel vehicles operated on the road. Data are then compared with data generated at emission test facilities to determine if current sampling and analysis methods are adequate for characterizing particle size and number. Then in order to determine the zone of influence of these emissions from a roadway, particle transformations are examined as the plume disperses downwind of the roadway in typical urban situations.

Accelerated Climate Prediction Initiative. The Accelerated Climate Prediction Initiative (ACPI, 1998), sponsored in part by DOE/OBER, responds to a need within the United States to produce the projections of climate variability and climate change necessary for the U.S. to participate in international assessments of climate change, as well as to understand the regional and national effects of global change. The promise of dramatic improvements in computational capabilities and simulation science affords the global modeling community an opportunity to make breakthroughs in understanding and projecting long-term changes in the global environment. Because of the radiative forcing of climate change due to anthropogenic aerosols and the need to represent this climate influence in climate models, it may be anticipated that aerosol modeling will be a component of the ACPI. The scientific understanding developed in TAP will be of direct and immediate relevance to that modeling effort.

4.7 Relation to Other Federal Programs

Because of the widespread recognition of the importance of tropospheric aerosols there is much interest in the several agencies responsible for air quality and global change in developing enhanced scientific understanding and model-based representation of the processes controlling the mass loading and chemical and microphysical properties of tropospheric aerosols. Several agencies have substantial research programs examining various aspects of the tropospheric aerosol issue. These are briefly summarized in Appendix C. Reference should be made also to the compilation of particulate matter research activities in the United States is maintained by the Health Effects Institute (HEI, 2001). In this context it is the explicit intent of TAP both to be complementary to these programs and to cooperate with these

programs to the mutual benefit of the several programs and the nation.

In view of the intense interest in tropospheric aerosols there is already strong interaction and cooperation among the several agencies at both the agency level and at the working scientist level. At the agency level, this interaction occurs in the Air Quality Research Subcommittee and in the Subcommittee on Global Change Research, and in the Executive Steering Committee of NARSTO, in all of which DOE plays an active role, as do the other cognizant agencies. At the working level, there are strong bonds that have been forged between DOE ESD programs and the corresponding programs in NOAA and EPA, as well as with researchers supported by NSF. These working-level interactions have been manifested in the past in jointly conducted field programs, most recently in the Southern Oxidant Study. There are also strong ties to aerosol research conducted in support of global change research by pertinent offices of NOAA and NASA that have been manifested in joint modeling activities and in active collaboration in use of ground-based and satellite data pertinent to aerosols. It may be accurately stated that these cooperative activities have led to a whole that well exceeds the sum of the parts in terms of the value of the research and even more so in terms of value per money expended.

It is wholly to be expected that connections such as these will continue into the future and even be strengthened. The problem of tropospheric aerosols is not only an important one; it is also a very difficult one. Consequently it may be anticipated that TAP Field Campaigns will be conducted jointly with field campaigns of other organizations, including states and the private sector, to the benefit of all parties. More specifically, as EPA establishes its several "supersites" for detailed characterization of aerosols and aerosol precursor gases (EPA, 2000), the presence of these sites will make these locations attractive for conduct of TAP Field Campaigns.

TAP interactions with the programs of other agencies will be further strengthened by participation on the Steering Committee of the TAP Science Team of scientists from the several other agencies. This participation will be especially valuable as the Steering Committee undertakes periodic reviews of the components of TAP in order to ensure that necessary gaps in knowledge are being filled, and if not, what new program components must be added to TAP. It may be hoped as well that the Steering Committee can work with corresponding entities in other Agencies, for example to ensure compatibility of methods, maximum utility and utilization of data, and sharing and intercomparing models.

5. Science Implementation

5.1 Field Measurements

TAP Field Measurements will be centered in intensive Field Campaigns of limited duration directed to the examination of aerosol mass loading and chemical and microphysical properties and to the evolution of these quantities through atmospheric processes. Abundant experience in studies of atmospheric chemistry has shown that only through field measurements can confidence be developed in the physical understanding and model representation of processes responsible for evolution of chemical substances in the atmosphere.

Each campaign will use a suite of sophisticated instrumentation deployed at an array of surface sites in combination with systematic in-situ measurements aloft by aircraft borne instrumentation and with remote sensing from the surface, aircraft, and satellites. These measurements will be supported by a network of sites that will provide meteorological data such as winds, mixing heights, and similar parameters necessary to characterize atmospheric transport.

Experience has shown that important advances in understanding of atmospheric chemical processes occur through the study of contrasts. Therefore field campaigns will be conducted in multiple locations, characterized by different climate, geography, and industrial and natural emissions, and at different times of the year. It is intended that TAP will conduct comprehensive field intensives at roughly one-year intervals.

In order to augment the measurements provided by TAP it is intended that TAP field campaigns take advantage of existing monitoring networks and specialized sites where measurements of particles and reactive gas concentrations are made, and of other field programs carried out by organizations having similar goals. In some cases it may be advantageous for TAP to join forces with the photochemical oxidant or boundary-layer meteorology community, thereby taking advantage of a common need for chemical and/or meteorological information. This approach clearly calls for a program that has the flexibility to conduct campaigns in locations in which other mutually contributing activities are taking place.

In addition to these large scale comprehensive field programs, there will be opportunities within TAP for smaller scale, more narrowly focused field studies. These might, for example, be directed at instrument evaluation, the testing of specific hypothesis, or supplementing the measurement capabilities of a long term monitoring site. Finally, TAP should be prepared to rapidly respond to major haze episodes, fires, or dust events, which can provide unique opportunities for studying aerosol processes. Studies during such events with support by forecasting, regional modeling, satellite monitoring, and continuous surface data will lead to rapid advances in understanding.

TAP field measurements will be centered in intensive field campaigns examining aerosol mass loading and chemical and microphysical properties and the evolution of these quantities.

TAP Field Campaigns

Major field campaigns will extend for typically 4 to 6 weeks, a time period adequate for the accumulation of a statistically sound data base that depends on sampling multiple 5- to 7-day synoptic cycles. This time period is also within the endurance time-frame for scientists and support staff to be in the field, and is affordable within anticipated resources. A further consideration governing the frequency of field campaigns is the necessity for time to analyze, interpret, and model the results from a given campaign, and to prepare reports of findings for publication in the scientific literature.

In the past, especially in the photo-oxidant research community, campaigns have

typically been conducted primarily during the summer, to take advantage of heightened photochemical oxidant activity as well as the availability of university researchers. However, it is necessary also to conduct campaigns during other seasons, for example, to study the winter "brown cloud" that is characteristic in certain urban regions.

A given field study will address multiple scientific objectives.

A given field study will focus on a geographical area of limited extent but will be designed to address multiple scientific objectives, for example aerosol evolution in power-plant plumes or plumes from isolated urban areas. Other scientific objectives might include examinations of the role of cloud processes in the formation of aerosol mass or the generation of new particles. Typically, field observations will be centered on an urban area. The spatial scale will extend from the urban center, the location of most intense emissions and photochemical activity to more regionally representative non-urban locations typically about 200 km. This spatial scale encompasses emissions of aerosol precursor gases, intense secondary photochemistry leading to gas-to-particle conversion and formation of new particles and growth of pre-existing particles. Study over this spatial scale also permits quantification of the export of aerosols out of the source region; this export is responsible for so-called regional background levels. Measurements conducted on such scales will also allow for targeted studies such as particle formation and growth in power plant plumes, cloud process studies, and aerosol removal by wet and dry deposition.

Field Campaigns will be centered on an urban area extending to more regionally representative non-urban locations, typically about 200 km.

A combination of airborne and surface-based measurements will be used. Figure 5.1 shows a schematic of the deployment of these measurement facilities. Shown in the figure are a central site ("supersite") at which the most detailed observations will be made. Measurements will be made by both in-situ and remote sensing instruments. This supersite will be augmented by several, typically 6, supplementary sites ("satellite sites") where a more limited set of measurements will be made to provide information on the spatial variation of aerosols and key precursor gases. Surface-based measurements will be supplemented by aircraft carrying in-situ and remote sensing instrumentation to provide information about the vertical and horizontal distributions of aerosols and precursor gases.

In addition to the chemical and microphysical information obtained by aircraft and surface measurements, it is essential to characterize the flow fields and vertical structure of the atmosphere to permit interpretation of the measurements. These measurements are also required as input to mesoscale models. A network of specialized meteorological instruments will be deployed to conduct such measurements. Additionally pertinent meteorological data will be acquired from external data sources.

Field Program Planning and Implementation

Planning and implementing a large scale and complex field program such as TAP is a complicated task. An inherent problem is providing sufficient and sufficiently well defined data to usefully address the broad range of scientific issues that need to be addressed by the program. Allocation of sufficient resources for field studies is a continuing problem.

A way of addressing these concerns that has been effectively used in other programs is the so-called "mentoring" concept, and it is suggested that this concept be used in TAP. In this model the broad scientific objectives of the program will be defined by the TAP Field Investigators Team. A subset of these scientists will then be selected as mentors to design and represent specific studies to be conducted as a part of the Field Campaign. Mentors will form teams of scientists with similar interests to specify the measurements, strategies for use of aircraft and similar factors that are required to achieve their objectives. The set of studies developed by the mentors will constitute the Scientific Plan for a given field program. During conduct of the program, the committee of mentors will be responsible for the day-to-day planning, converting the measurement requirements into a set of plans that best uses the deployable resources such as aircraft and surface measurement facilities under a given set of meteorological conditions. Mentors will also be responsible for initiating and coordinating subsequent data analysis. A

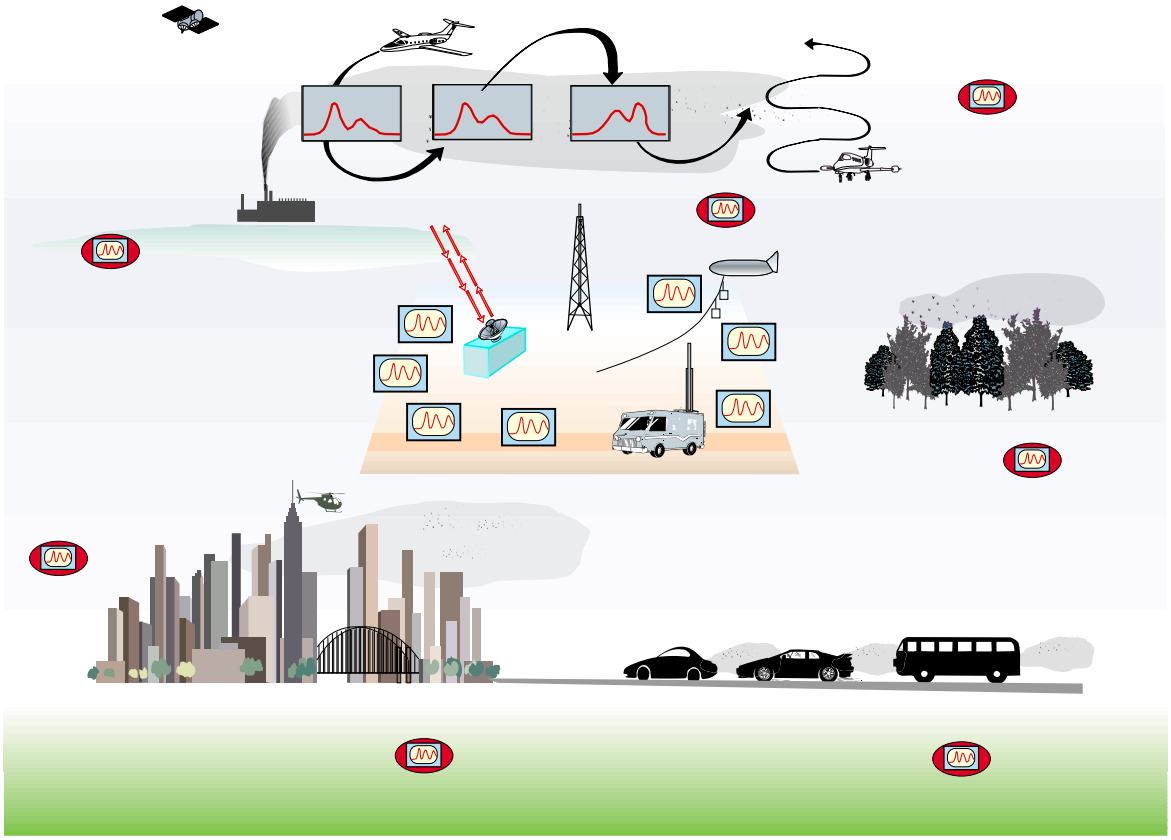


Figure 5.1. Schematic Diagram of TAP Field Campaign. Supersite (center) provides detailed characterization of aerosols, precursor gases, and meteorological variables by in-situ measurements and remote sensing continuously over the 6-week campaign. An array of satellite sites provides continuous information on the spatial variation of these quantities, but with less detail. Measurements with fixed-wing aircraft and helicopters provide in-situ and remote sensing information on the vertical and horizontal distributions of these quantities. Satellite observations provide an overview of the large scale distribution of aerosols and synoptic-scale meteorology.

field program manager will be responsible for overall coordination of the Field Campaign, overseeing the planning process, and procuring resources for the study, and will preside over the campaign itself. An important function of the field program manager will be to assure that all of the mentors obtain an adequate share of the available resources during the campaign.

Measurement Strategies

A detailed set of measurement strategies will be developed for each field program and summarized and made available to TAP investigators and other participants in the form of a Field Program Science Plan. Specific strategies will be determined by the set of objectives to be addressed in a given program. Defining how, when and where these studies will be conducted will be the responsibility of the TAP Field Investigators Team (see Figure 4.1).

The overall strategy of TAP field program design will be to provide a structure and basic set of measurements so that the specific objectives of a given Field Campaign can be achieved. At minimum it is anticipated that TAP will provide a continuous description of the concentration and composition of aerosols, aerosol precursors and other relevant species,

as a function of time, location and altitude within the project area as an integral part of each Field Campaign.

This description of aerosols and precursor gases will be accompanied by an equally detailed characterization of the meteorology. In addition to mean winds the measurements must include turbulent fluxes, boundary layer growth, fair weather cloud formation, cloud evolution and microphysics, vertical transport and mixing processes, differential advection of temperature and humidity, atmospheric stability, and entrainment. Although TAP measurements will typically be limited to a 200-km domain, it will be necessary to take into account the influence of synoptic-scale processes on transport of aerosols and aerosol precursors into and out of the study area. These baseline data are required both to provide the context within which measurements can be interpreted and to drive and evaluate mesoscale models of aerosol loading and transport.

The description of aerosols and precursor gases will be accompanied by an equally detailed description of the meteorology.

A strategy for deploying surface sites that has been used successfully in field campaigns examining photochemical oxidant formation is to perform detailed measurements at a primary site and to augment these measurements by a network of satellite sites. This is the approach that will be taken in TAP, as indicated schematically in Figure 5.1.

The TAP supersite will deploy a comprehensive suite of instruments that will give rise to a comprehensive set of measurements of aerosols, aerosol precursor gases and other controlling or diagnostic atmospheric chemistry variables, such as actinic flux. This suite of instruments will be centered on one-of-a-kind devices that extend the state of the measurement art or which are so scarce or costly to use, that they can be deployed only at a small number of sites. Baseline measurements at these primary sites will include O_3 and O_3 precursors such as NO, NO_2, NO_y, and volatile organic compounds, VOCs; aerosol precursors such as SO_2; photochemical intermediates such as formaldehyde, and product species such as HNO_3 and peroxides. Aerosol characterization measurements will include microphysical properties (e.g. size distributions from 3 nm to 10 μm), coarse and fine bulk composition, size-resolved composition, aerosol mass, and multi-wavelength aerosol light scattering and optical thickness. Examples of specialized instruments include single-particle mass spectrometers, Lidars for measuring the vertical distribution of aerosols and water vapor, and mass-spectrometric instruments tailored to the measurement of gas-phase H_2SO_4 and NH_3. These measurements will be supported by a full complement of meteorological measurements including radio-acoustic sounder wind profilers.

Satellite sites will necessarily have less extensive measuring capabilities, but as they will be less costly to establish and operate, they can be placed in more locations. These sites will be used to measure the most important gas-phase species and provide a coarser description of aerosol composition and size distribution. Where possible TAP will make use of aerosol and gas measurements from air quality measurement sites maintained by state or regional agencies.

A possible siting strategy for TAP will be to locate the supersite immediately downwind of an urban center to characterize the emissions from the city into the downwind domain and to study fast conversion processes. Satellite sites will be located at several upwind and downwind distances and/or at locations in chemically different environments (e.g., coastal, forested) within a 200 km measurement domain. The number and disposition of sites within a project area will vary from campaign to campaign, as each study location will have a unique set of characteristics that will need to be addressed in the field campaign design.

Essential to many process level studies are the time variation of winds and boundary layer height needed to determine dilution and to infer the time scale over which reaction and aerosol evolution have occurred.. These data are also required for driving regional scale models used to calculate concentration fields. To meet these requirements a network of meteorological instruments will be deployed to provide information on wind fields, boundary layer height and other important parameters. Wind fields will be measured with a network of

Radio Acoustic Sounding Systems (RASS). These will be supplemented by periodic release of radiosondes. The wind profilers will be located at sites where chemical and aerosol properties are also measured, thereby enhancing the value of both measurements. A particularly useful set of measurements will be obtained by co-locating a RASS with a Lidar measuring aerosol vertical profiles. This combination will provide the meteorological context needed to interpret variations in aerosol vertical profiles and the concurrent surface- and aircraft-based measurements of aerosol properties. The number and disposition of meteorological instruments for each of the Field Campaigns will be depend on the location in which the campaigns are conducted. Locations which exhibit complex meteorology (e. g. coastal locations with sea-breeze circulation or locations in complex terrain) will require a fairly dense network of measurements. Locations with relatively simple meteorology will require fewer sites.

An example of a deployment strategy for surface sites that may serve as a useful model for TAP Field Campaigns is that used in the Southern Oxidant Study Summer 1999 Nashville Intensive, Figure 5.2. The focal point of the surface network (corresponding to a TAP "supersite") is located just to the northeast of downtown Nashville, where an extensive set of instrumentation permits detailed characterization of the rate of ozone formation and the important factors controlling this rate. The site was selected to be at or near the point of maximum ozone formation under prevailing wind conditions. A site in downtown Nashville is used to document the urban source signature. An additional site

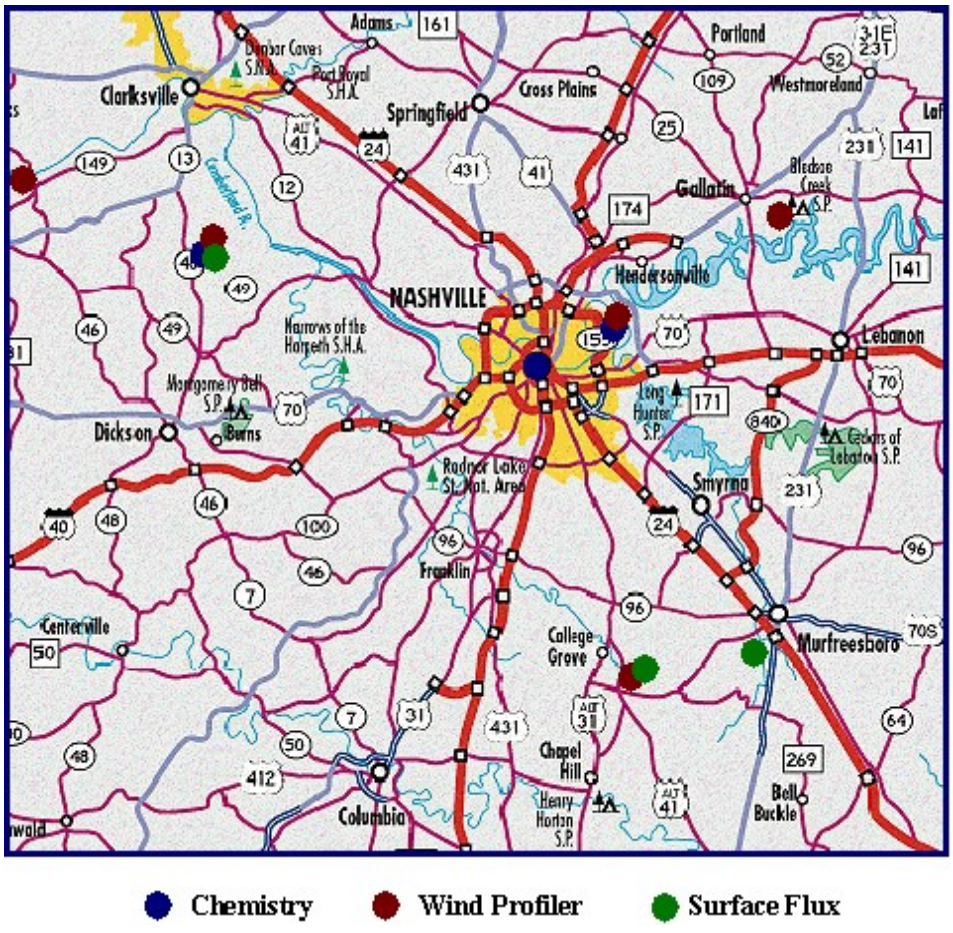


Figure 5.2. Deployment of surface sites for measurement of chemical constituents, wind flows, and surface fluxes in the Southern Oxidants Study 1999 Nashville Experiment. From SOS (1999).

northwest of the city (in the prevailing upwind direction) characterizes the background air.

Similar considerations will guide the placement of sites in TAP except that the principal focus will be aerosols rather than photochemical species, and the time/distance scales may be somewhat different.

Instrumented research aircraft will be an integral component of TAP Field Campaigns. They will perform a variety of studies not possible with either surface-based or remote-sensing measurements.

Instrumented research aircraft will be an integral component of TAP Field Campaigns. They will perform a variety of studies not possible with either surface-based or remote-sensing measurements. Examples include particle formation and evolution in plumes, cloud process studies, regional characterization measurements, and optical closure experiments. Use of multiple aircraft is planned. A slow-flying fixed wing aircraft or helicopter is needed for measurements of source characteristics. A large aircraft equipped with a full complement of instrumentation for measurement of aerosol precursors, intermediates, and product species, and aerosol and cloud microphysics will be needed for plume studies, cloud process studies, and regional characterization measurements. The in-situ measurements of these aircraft will need to be supplemented by an aircraft equipped with instrumentation for remote sensing of quantities such as the vertical distributions of aerosols, water vapor, and photochemical species such as O_3. These measurements will allow quantities such as the vertical and horizontal dispersion of plumes from urban and point sources to be determined. In the past, coordinated remote-sensing and in-situ aircraft measurements have been extraordinarily valuable, with the remote sensing measurements providing a context within which the in-situ measurements are interpreted.

Detailed strategies for deployment of aircraft and other resources will be developed by the TAP Field Investigators Team as part of the planning activity for each Field Campaign. The following paragraphs present examples of strategies that have been used in previous studies and which will form a basis for developing measurement strategies in TAP.
One previously employed strategy for effectively studying oxidant formation has been urban plume studies, and it is anticipated that many features of aerosol formation and growth can also be studied in urban plumes. Aircraft are the ideal platform for such studies owing to their ability to rapidly sample large distances. Sampling would be done upwind of the source region, over the source region, and in the plume emanating from the source at various times (distances) downwind. An example of such a flight pattern used to study the evolution of the chemical and microphysical properties of several power plant plumes during the Summer 1995 Southern Oxidant Study Nashville Intensive is shown in Figure 5.3.

The upwind and source region flight segments serve to quantify inputs into the plume from background air and from the source itself. Wind speed is used to convert a known downwind distance to an atmospheric reaction time, thereby permitting the observed changes in aerosol properties to be expressed in terms of rates. Of particular interest are the rates of gas-to-particle conversion of various species, how this conversion adds to aerosol mass, and associated changes in the particle number and mass size distributions. The measurements made on the aircraft will be used to relate these rates to gas-phase processes.

Although such studies can in principle be done in a Lagrangian framework, in which a specific air parcel is intercepted at various downwind distances, consideration of the time and distance scales suggests that a true Lagrangian study would be operationally very difficult to achieve. Under favorable conditions a plume from an urban or power plant source can be tracked for about 200 km. For a typical boundary-layer wind speed of 20 km h^{-1}, measurements over a 200 km distance would allow chemical and microphysical changes to be tracked for the important initial 10 hours of the material's lifetime in atmosphere. A true Lagrangian study encompassing such a time span would require measurements made over a 10 hour period, considerably greater than aircraft endurance will allow. Multiple flights and/or multiple aircraft can be used to bypass this sampling incompatibility. However, experience by the photochemical community tracking urban and power plant plumes

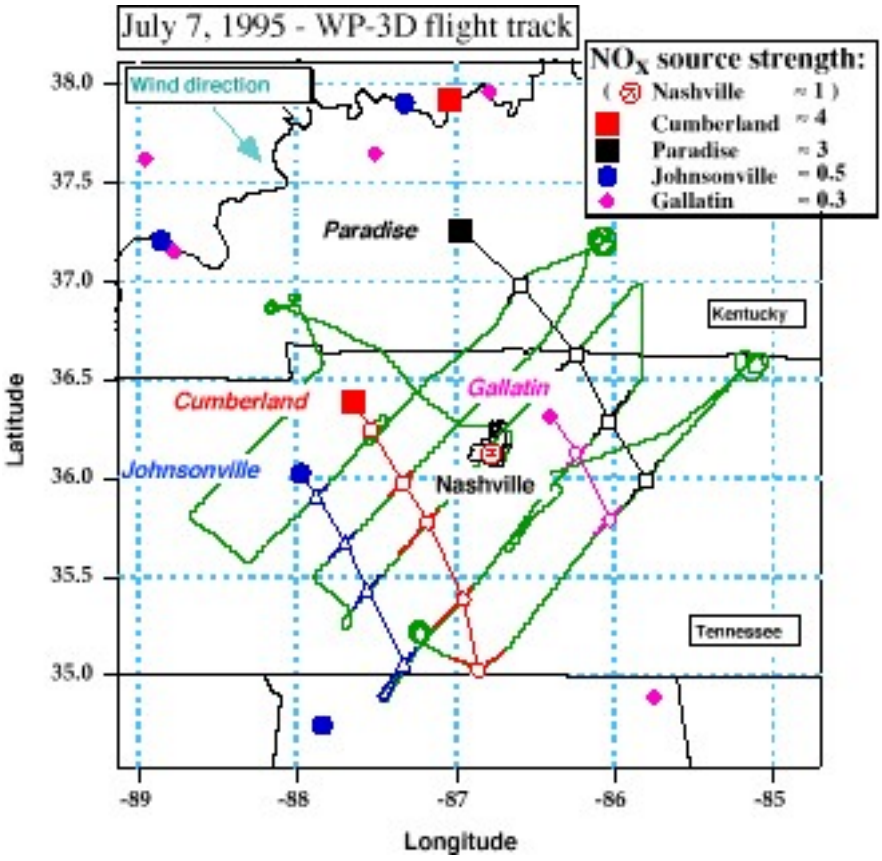


Figure 5.3. Example of typical flight path executed by research aircraft in the study of oxidant formation in power plant plume in the vicinity of Nashville TN. The color code identifies the flight track of the aircraft (green) and the various point sources in the region, the direction of their plumes and the plume-crossings by the aircraft. The estimated relative magnitude of the various sources of NOx are listed in the box. From SOS (1998).

suggests that not much information is lost by operating in a quasi-Lagrangian mode, in which the requirement of sampling in the same air parcel is relaxed. Plume traverses can then be done on air parcels that passed over the source region at any time within a several hour period. An example of the evolution of aerosol size distribution from a power plant plume study using a similar strategy is shown in Figure 5.4.

Development of a quantitative understanding of aerosol formation in urban and power plant plumes will be greatly enhanced by the addition of two additional elements, concurrent release of an inert tracer, and synchronized (to the in-situ aircraft) overflights by an aircraft with the ability to remotely sense the vertical distribution of aerosols. Release of tracers concurrent with the experiment allows a much more quantitative assessment of the rates of transformation processes to be developed, as losses of substances from the plume, and dilution can be accounted for by ratioing concentrations to the concentration of the inert tracer. The technology for releasing tracers from such point sources and for measuring them from aircraft platforms is well developed and available for use in TAP. (Koffi et al., 1998).

Over-flights with a remote sensing aircraft and passive sensing of aerosols by geostationary satellites provide essential contextual information for interpretation of the in-situ data. Because in-situ aircraft measurements provide only sparse sampling of a feature such as a plume, there is generally no way of knowing

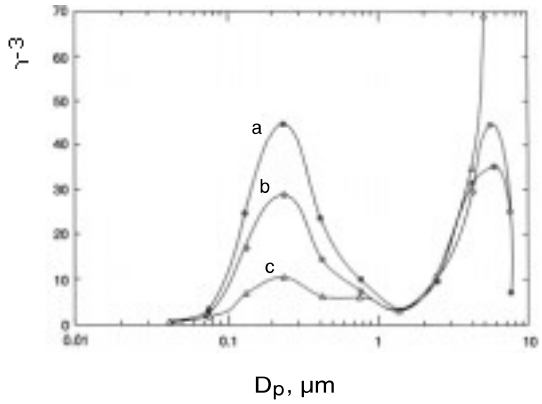


Figure 5.4. Aerosol volume-size distribution at 16 km (a) and 23 km (b) downwind in the plume of the Cumberland power plant in Tennessee, after background subtraction, and in background air (c). Note growth of volume in size 0.1 to 1 μm. Source McMurry et al., 1981.

from such measurements which portion of the plume is being sampled. As a result it is not possible to infer the influence of spatial inhomogeneities on aerosol evolution. The resultant uncertainties can be greatly reduced by using information on the context of the in-situ measurements relative to the plume as a whole. Such measurements can be made with a down-looking aerosol Lidar on a second aircraft overflying the plume.

Different strategies will be used to study other processes such as the interactions of clouds and aerosols. Clouds play an important role in determining the physical and chemical properties of ambient particulate matter. Particles increase their mass and change their composition and physical properties through in-cloud gas-to-particle conversion processes associated with aqueous-phase reactions. Cloud droplet aggregation and precipitation exert major influences on the evolution of aerosol size distributions. New particles can be formed near the tops of clouds when pre-existing aerosol surface area is depleted and photochemically active gas phase precursor concentrations are high.

Sampling strategies will be developed to test emissions inventories.

Cloud sampling (other than for fog) will be accomplished by in-situ aircraft sampling. The generalized cloud sampling strategy will be to characterize the trace gas, aerosol composition, and microphysical properties of air in the cloud inflow region, measure the composition of cloud water and cloud interstitial air, and to characterize the microphysical properties of the cloud at various altitudes within cloud. The chemical and microphysical properties of the air in the cloud outflow region will also be measured. Analysis of the relationships between the properties of the inflow air, cloud water and interstitial cloud air composition and microphysical properties, and similar properties in the cloud outflow air will provide key information on the role of clouds in modifying aerosol properties. Such flights may also provide key information regarding the conditions under which new particle formation occurs. Cloud studies would greatly benefit from measurements with co-located cloud radars, which can define the physical boundaries of the sampled clouds, as well as provide information about cloud dynamical processes that are key to understanding cloud-aerosol interactions.

Sampling strategies will also be developed to test emissions inventories. Emissions characterization, including the continued development of emissions estimation systems, continues to be a critical element of reliable air quality modeling, but generally has received low priority in apportioning resources in field campaigns. Further progress in understanding of tropospheric aerosols through interpretation of models depends critically on substantially improving emissions estimates. In the simplest form, these tests will be performed by comparing observed ratios of aerosol components with inventory predictions. This comparison is one of the few ways in which emission inventories can be tested. Testing and/or development of emissions inventories will require the development new methods to efficiently obtain, by analysis of data and activity patterns, emissions information on daily to hourly time scales commensurate with the requirements of the physical and chemical processes taking place in the study zones. Emissions data for stationary sources of sulfur oxides, nitrogen oxides and VOCs will be

needed along with transportation data. An equally important need will be ammonia emissions data, from sources quite different from the others, including natural and agricultural categories.

Measurements and Platforms

The instrument suite that will be employed in a given TAP Field Campaign will be determined during the campaign planning process. To a great extent the availability of instruments and their operational characteristics such as time response and sensitivity will determine field measurement design. Some of the required instruments are available, are well calibrated and have operational characteristics that are well understood. Other instruments will need to be improved so that their time response, sensitivity, or specificity are adequate for their intended use, e. g. aircraft deployment. Some instruments will need to be developed and characterized before they are deployed in the field. A listing of state of the art instrumentation suitable for deployment at either surface sites or on airborne platforms is given in Appendix B. A key inadequacy is measurement of organic composition. Promising techniques have been identified, so that it may be expected that new instrumentation can be integrated into TAP Field Campaigns on a relatively short time scale. There will be a strong linkage between Instrument Development and Characterization investigators and the Field Studies investigators to define the measurement needs so that development activities directly meet both the scientific and operational needs of the program.

In addition to aerosol properties, TAP Field Campaigns will also require measurement of various trace gas species that are aerosol precursors (e.g., SO_2, NH_3, organics, etc.) and species that characterize the photochemical milieu in which aerosol transformations occur. It is anticipated that at least one of the aircraft utilized in the program will need to carry a full complement of instrumentation to characterize the photochemical properties of the atmosphere in addition to instrumentation for characterizing important aerosol properties. Other aircraft involved in the program will need to be equipped with a subset of this instrumentation. The species that must be measured include photochemical precursors such as NO_x and hydrocarbons, intermediates such as aldehydes, and product species such as O_3, HNO_3 and peroxides. Similar detailed measurements will need to be made on the surface at the supersite and at least one satellite site. Less detailed measurements, perhaps including only NO_x, O_3, CO, and hydrocarbons will need to be measured at other satellite sites. In many locations contemplated for these Field programs, the need for measurements at satellite sites can be met with data from existing networks operated by state and federal agencies.

The instrument suite to be deployed in a given TAP Field Campaign will be determined during the campaign planning process.

The regional approach of TAP and the focus on the life-cycle of ambient aerosol dictates that consideration be given to the different types of mobile sampling platforms best suited to accomplishing TAP's mission. Mobile sampling platforms include large and small aircraft, helicopters, airships, balloons, and tethered kitetoons. The focus of TAP on aerosols and their precursors from the surface to the tropopause over horizontal scales of 200 km requires cross-disciplinary, multi-instrument aircraft payloads.

Field campaigns will require aircraft having different sampling and measurement characteristics. For studies of aerosol transformations in urban or power plant plumes, the use of a helicopter to determine source characteristics is invaluable. Urban source regions are only marginally accessible by conventional aircraft owing to the usual air traffic control restrictions, whereas helicopters can easily fly in these regions using patterns and altitudes that enable good characterization of the source region. Also, their lower cruise speed allows spatially resolved measurements to be made during traverses of compact plumes such as those from isolated power plants. Helicopters were effectively used during the Summer 1995 Southern Oxidant Study Nashville Campaign to characterize the Nashville urban source region and to conduct studies of power plant plume chemistry.

Studies of plume chemistry downwind and general characterization of aerosol properties throughout the project area will require a large

aircraft fully equipped with the required complement of aerosol/photochemical characterization instruments. Because of the large amount of instrumentation required on a single platform, this aircraft will have to have significant payload capacity and be capable of fairly long duration (4 to 6 hr) flights. Aircraft that meet these requirements include the DOE Grumman Gulfstream G-1, the NOAA P-3, the National Center for Atmospheric Research C130, the Canadian National Research Council Convair 580, or the University of Washington Convair 580.

TAP Science Support will provide and operate the platforms and baseline instruments and make meteorological measurements during Field Campaigns.

Campaigns will also require the use of an aircraft with the capability for remote sensing of aerosols to define, for example, the vertical and horizontal aerosol concentration fields emanating from a source, or to characterize the concentration fields of aerosols within the project domain. Because the flight requirements for the remote sensing aircraft will generally be different than the aircraft used for in-situ measurements, a separate aircraft will be necessary. In several previous field programs remote sensing aircraft have flown in stacked configuration with the in-situ aircraft necessary to provide a context for interpretation of the in situ measurements. The remote sensing aircraft could also be used for other optical measurements of aerosol properties, and potentially for cloud physics measurements. Potentially a number of aircraft could fill this role including the Construcciones Aeronauticas SA (CASA) 212 operated under contract for remote sensing to the NOAA Aeronomy laboratory, or the University of Washington Convair 580.

Science Support Requirements

Support necessary to conduct field campaigns as outlined in each Field Campaign Science Plan will be provided by TAP Science Support. This effort will include preparation of Operations Plan for a given campaign, consistent with Science Plan, that will allow these questions to be addressed using available technology and resources.

It is expected that TAP Science Support will be responsible to provide and operate the platforms and baseline measurement instruments during Field Campaigns. The platforms will include three aircraft, one for source sampling, one for comprehensive chemical and physical measurements and one for remote sensing; the TAP supersite, and 3 to 6 satellite sites. It is anticipated that the remote sensing aircraft and the helicopter will come equipped with instrumentation required for their respective tasks. TAP Science Support will be responsible for measurements that utilize tested and established methods, as distinguished from Principal Investigator instruments, which will be supported through peer-reviewed funding process. It is expected that Science Support will, at minimum, provide quality assured measurements at the surface sites and by aircraft of gas-phase species such as SO_2, NO, NO_2, NO_y, and CO; accumulation-mode aerosol size distributions; filter collection and analysis for major aerosol components; aerosol mass (surface only), and related quantities.

TAP Science Support will also be responsible for providing the basic meteorological data for these studies. This will include meteorological soundings (rawinsondes), a detailed description of wind fields and boundary layer height (profiler network), and weather forecasting and nowcasting to assist in Campaign Operations. Operation of chemical transport models in real time and forecast mode will enable most effective deployment of TAP resources, especially aircraft platforms.

TAP Science Support will also have major responsibility for obtaining external data during Field Campaigns and afterwards in the analysis and interpretation of Campaigns. An important example is obtaining satellite data. Satellite observations can be used to define widespread regional aerosols (Lyons, 1980). Such data will provide important context for TAP measurements and therefore need to be made available in real time for planning and conduct of the field measurements. Quantitative aerosol retrievals are available from a number of current satellite instruments, albeit with limitations due to frequency of overpass and interferences in measured radiance (King et al., 1999). Considerably improved aerosol products will become available following launch of the NASA Earth Observing System AM platform, expected in

late 1999 (EOS, 1999). Next-generation satellite products will be used for their unique synoptic viewpoint of aerosol column burden, and, in the near future, vertical structure by satellite-borne Lidar.

The Science Support Team will also have the responsibility to construct the deployable supplemental facilities, locate and obtain suitable sites, typically in conjunction with local air quality entities or educational institutions, provide communications, office space for participants, lab space as required and a user-friendly mechanism to allow ready accessibility of data by all participants during campaigns and the resources to archive the data for subsequent analysis. Other support functions include QA/QC and data base maintenance. TAP management will assist PIs in disseminating scientific results by providing for press releases, workshops, conference sessions, and special issues of journals.

5.2 Instrument Development and Advanced Characterization

Atmospheric aerosols consist typically of an external mixture of particles that are themselves composed of a multitude of internally mixed compounds. For TAP activities to successfully quantify the chemical and physical properties of ambient particles, this complexity must be recognized and determined. Improved understanding of source-receptor relationships and the potentially harmful health effects of ambient particles requires information on the size-dependent aerosol chemical composition Many of the compounds comprising aerosol particles are volatile or semi-volatile (such as ammonia, nitric acid, and organics), resulting in exchange with the gas phase while in the atmosphere as well as in artifacts in sampling and analysis. TAP field measurements will therefore require reliable, calibrated, and well characterized instruments to measure the physical and chemical properties of ambient particles and the air in which they are embedded. The Instrument Development and Advanced Characterization (IDAC) component of TAP will be closely linked with the field component and participants in the instrument development component will participate in the field programs.

Traditionally, aerosol studies have focused on classifying aerosols according to their size, number concentration, and bulk chemical composition; relatively few detailed measurements of size-resolved composition have been made. A comprehensive suite of measurement capabilities will be required to address the scientific questions of TAP. Table B.1 in Appendix B lists capabilities for in-situ measurements of aerosol physical and chemical properties that are currently available or are expected to be available at the outset of TAP. The list is not intended to be exhaustive, however, it demonstrates that many different measurement techniques must be integrated in order to obtain a more complete picture of the gas/particle equilibrium.

Key areas of instrument development include measurement of aerosol chemical composition, characterizing aerosol physical properties, and integration of physical and chemical property measurements.

The key areas where development of new instrumentation or refinement of existing measurement techniques is required to address the TAP research tasks include measurements of aerosol chemical composition, characterizing aerosol physical properties, and integration of physical and chemical property measurements. Related to these key areas, TAP should focus instrument development on techniques for quantifying the composition of the organic fraction of ambient particulate matter, measurements of the gas-phase species that form new particles, size-resolved chemical composition measurements, and integration of physical and chemical measurement techniques. For each of these areas, rapid-response aerosol measurement techniques may be required, particularly for aircraft platforms and to observe how certain physical and chemical processes affect ambient particles.

Aerosol Chemical Composition

The chemical composition of the atmospheric aerosol varies with particle size, geographical location, and time. Particles with the same

diameter have also been shown to exhibit different chemical compositions. Ultimately, determining which sources of aerosols must be regulated requires understanding of the variation of chemical composition with particle size from the sub-1 nm molecular scale to 10 μm diameter coarse particles. Size-resolved composition measurements can provide information on mechanisms of chemical transformation and on the contributions of various species to particular aerosol effects. New instruments are required to improve understanding of atmospheric aerosol chemistry, particularly in quantifying the variety of species that are present, observing the chemical evolution of the aerosol, and measuring aerosol formation processes.

Single particle analysis is required to determine size-resolved chemical composition of ambient aerosols and the state of mixing of aerosol constituents.

Organics are virtually always present in atmospheric aerosols. They can comprise a significant fraction of the total aerosol mass (often between 20 and 50%) and may play an important role in new particle formation. Current state-of-the-art techniques are capable of speciating only between 10% and 20% of the organic fraction of the ambient aerosol. Understanding the organic aerosol fraction is thus a high priority.

A major effort is needed to develop real-time methods to measure the polar organic component of particulate matter, as these compounds are expected to play an important role in determining the water uptake properties of organic containing particles. The photochemical evolution of the organic aerosol can also be investigated by measuring the changing degree of oxygenation of the organic aerosol. Useful measurements of this type include: organic mass fractionated by polarity, measurements of functional groups associated with organic particulate matter, measurement of selected molecular-level tracers of primary and secondary organic carbon, and measurements of water-soluble and water-insoluble organics. Continued development of these types of measurements would be extremely valuable.

The lack of closure between measured organic species and gravimetric mass, especially for situations with a significant organic loading, could be due to an inaccurate value for the average molecular weight per carbon weight. Thus, measurements leading to a more accurate assessment of this quantity are necessary.

The above techniques for quantifying organic species present in the ambient aerosol must be capable of providing some particle size-resolved information. Techniques should be capable of discriminating primary from secondary organic particulate matter and between anthropogenic and biogenic organic carbon. Semi-volatile and water soluble organics can be either desorbed or absorbed during collection, and instrumentation that minimizes these sampling effects is required. Instrumentation developed for aircraft platforms must be as small and light as possible to facilitate deployment.

Nitrogen containing aerosols, another major component of ambient particulate matter, are known to be an important contributor to secondary aerosol loadings. Much of this material is derived from the transformation processes involved in the oxidation of nitric oxide emissions from combustion sources and the reaction of ammonia with acidic gases and aqueous aerosols. Measurement methods for ammonia are needed with time resolutions from seconds to minutes and detection levels between 10 and 30 ppt. Organic nitrogen compounds are of concern due to their abundance, potential toxicity, radiative properties, and for acting as a sink and source of NO and nitrogen dioxide. These compounds have received considerably less attention than the corresponding inorganic compounds, and instrumentation to allow rapid and sensitive detection of organo-nitrogen species is needed for both ground and air-based field measurements.

Single particle analysis techniques are required to provide unique information on the size-resolved chemical composition of the ambient aerosol and the state of mixing of aerosol constituents (internal vs. external); these properties can be used to identify sources and growth mechanisms of aerosols. Techniques that can be used to analyze individual particles can be divided into those that require later laboratory analysis of samples from the field and those that analyze particles in real-time. For example, transmission electron microscopy (TEM) can provide unique information on the size, shape,

mixing state, and composition of individual particles. Figure 5.5 shows examples of single particles of mixed composition of seasalt and sulfate and nitrate. These measurements can be made with sufficiently high time resolution to permit aircraft sampling, as a statistically meaningful number of particles can be collected in a short period of time. The particles are subsequently analyzed in the laboratory. Major improvements in TEM instrument design are nonetheless required, including automation of the measurement process, interfacing and comparing TEM measurements with other measurement techniques, developing the capability to speciate organics, and instrument calibration.

In contrast to the TEM technique, which is performed "off-line", time-of-flight mass spectrometric (TOF-MS) techniques can provide real-time information on trace species present within individual particles. Examples of the complex spectra resulting from the TOF-MS analysis of three different particles are shown in Fig. 5.6. Measurements such as these demonstrate that individual particles can consist of internal mixtures of several different chemical species. TOF-MS techniques are not currently capable of providing quantitative real-time information on mass concentrations, and improved quantitative capability is an important area of development for these instruments.

New single-particle techniques are required for speciating the wide range of organic compounds. Similarly, there is a need for surface selective analysis capabilities to evaluate, for example, the role of the particle surface chemical composition in determining hygroscopic properties. TOF-MS systems also need to be made cheaper, smaller, and with higher sampling efficiencies. Advanced data processing methods are required for efficiently analyzing and interpreting the large amounts of data that are generated.

The focus of TAP on the processes controlling the life-cycle of the ambient aerosol requires that instrumentation be available to measure the chemical evolution of particles and precursor gases. Instrument packages are required that can distinguish and examine primary particles, i.e., particles emitted directly

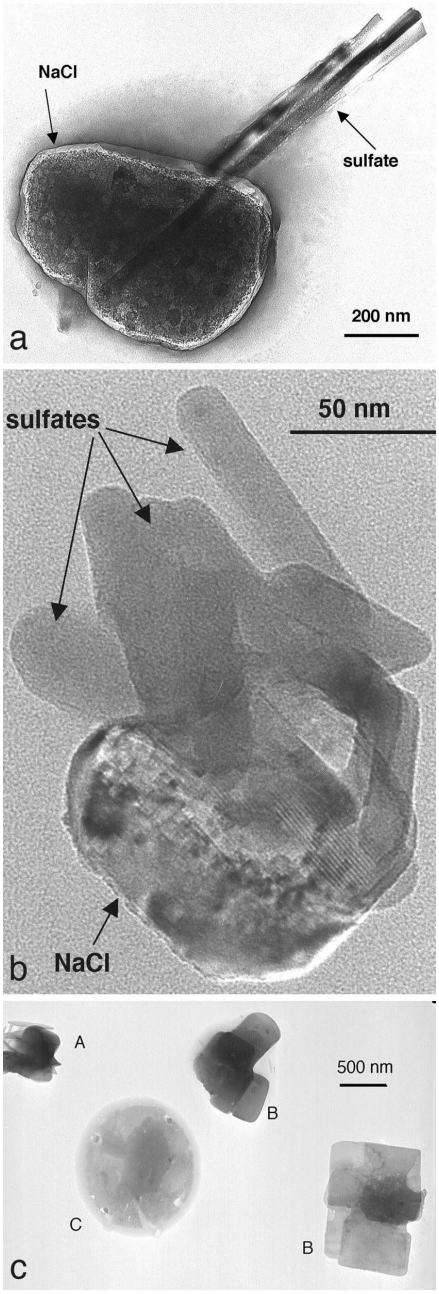


Figure 5.5. Examples of Transmission Electron Microscopy (TEM) images of single aerosol particles showing detail of compositional and size information that may be gained. Images a) and b) show subhedral halite (NaCl) and euhedral sulfate crystals. The particle in b is in the smallest size range of seasalt crystals. The images in c show halite particles in various stages of conversion to sulfate and nitrate. Grain A is partly converted, whereas C has been completely converted to nitrate and grains B to sulfates. From Buseck and Pósfai (1999).

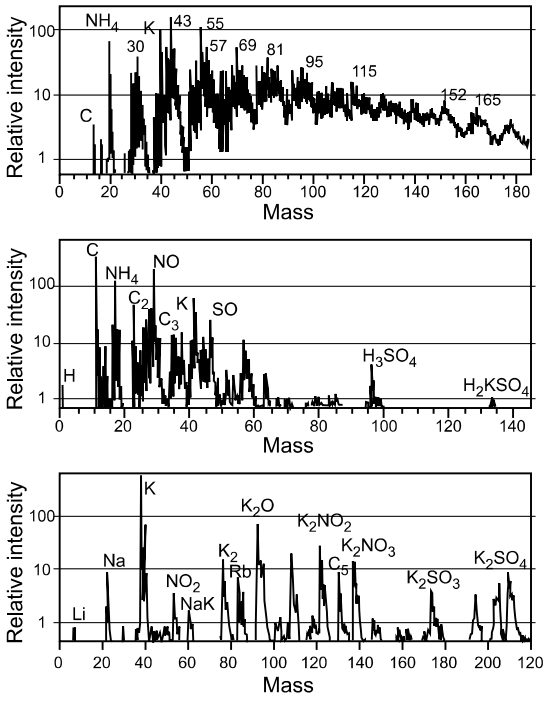


Figure 5.6. Positive ion mass spectra of several single particles Idaho Hill taken during the month of September 1995. Note substantial particle-to particle variation in composition. Particle 1 is largely organic, with small contributions from inorganic ions. Particle 2 exhibits a mixed composition organic and inorganic. Particle 3 consists primarily of inorganic species including nitrates sulfates and a large variety of counter ions. From Murphy and Thomson (1997).

Methods are needed to distinguish primary particles, i.e., particles emitted directly by sources, from secondary particles, produced by gas-to-particle conversion in the atmosphere.

from sources, from secondary inorganic and organic particles, produced via gas-to-particle conversion in the atmosphere. In terms of primary particles, there is currently no established technique for measuring the contribution of mineral dust to the coarse particle population. Elemental carbon (EC) and organic carbon (OC) are found as primary aerosol, and measurement techniques for both suffer from a lack of calibration standards.

There are currently no total particulate mass measurement techniques suited for aircraft platforms, making mass closure studies extremely difficult to perform with aircraft data. For secondary inorganic aerosols, instrumentation detection limits must be decreased to below 0.1 $\mu g\ m^{-3}$ to successfully differentiate secondary from primary aerosols during chemical evolution studies. Secondary organic aerosol evolution needs to be examined within the context of the primary organics. More specific organic compounds in the organic fraction of PM2.5 need to be identified to determine which are definitely primary, which are secondary, and which ones derive from both processes. In addition, continuous measurements of heavy hydrocarbon (>C_8) precursors and oxygenated gaseous end-products are required.

In summary, a variety of chemical measurement techniques are required that can quantify the bulk and size-resolved chemical composition of ambient particles. Instruments are needed that can measure the composition of individual particles and the surface composition of particles. A variety of chemical species must be measured by these systems, including inorganics, organics, trace metals, and soot. In order to evaluate the fraction of the total aerosol mass that has been quantified by the chemical measurements, and to place the chemical composition measurements in a more regulatory context, simultaneous measurements of the total aerosol mass are required.

Aerosol Physical Properties

Particle properties such as shape, density and refractive index determine how particles are size-classified by measuring instruments. For example, impactors and aerodynamic particle sizers classify particles according to aerodynamic size which depends on size, shape and density; while optical particle counters measure an optical equivalent size which depends on size, shape and refractive index. Electrical mobility classifiers segregate particles according to electrical mobility which depends on shape. Furthermore, many atmospheric particles are hygroscopic, and because their water content, index of refraction, and density depend on relative humidity, their physical-chemical and optical properties do as well. This dependence needs to be understood to quantify aerosol measurements and to understand certain aspects of aerosol behavior. Several

approaches for measuring particle shape, density and refractive index that are suited for laboratory-generated aerosols have been reported, but uncertainties that arise from the complexities of physical and chemical morphologies of real atmospheric particles are typically not well established.

A critical need that must be met to improve current understanding of the processes leading to new particle formation is measurements of gas-phase precursors like sulfuric acid, ammonia, and organics. Measurements of these precursors, when conducted simultaneously with both the pre-nucleation molecular cluster size distribution and the pre-existing aerosol size distribution, will allow the conditions leading to new particle formation to be defined. Furthermore, some knowledge of both the bulk and individual chemical composition of particles in the 3-4 nm size range is needed so that relevant gas-phase precursors can be unambiguously associated with newly formed particles.

There is an important need for instruments to identify the presence of newly formed particles and to quantify the rate of formation and growth of new particles. A recently developed technique involving pulse-height analysis is capable of determining the size distribution of recently formed particles in the size range between 3 and 10 nm but requires refinement for application in urban-influenced areas. Representative results from the field deployment of the pulse-height unit are shown in Fig. 5.7, where the growth of newly formed particles is clearly evident.

The particle number size distribution is a fundamental physical property required for understanding the evolution and gas/particle partitioning of the ambient aerosol. In urban areas, significant variability in the number size distribution often occurs over time-scales shorter than a few minutes. Furthermore, for aircraft platforms, size distribution measurements are required over time-scales of a few seconds.

Ambient particle size distributions are made up typically of three or four modes. The chemical composition of these different modes, and therefore their hygroscopic properties, can

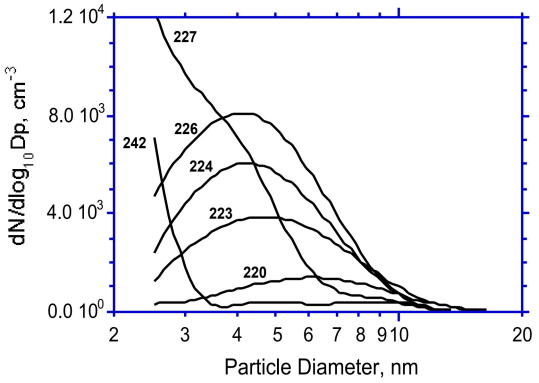


Figure 5.7. Selected nanoparticle size distributions measured far downwind of Macquarie Island. The sequence of measurements with increasing downwind distance is 242, 227, 226, 224, 223, and 220. The distributions show growth of newly formed particles to larger sizes close to the island and then depletion of nanoparticles with increasing downwind distance. From Weber et al. (1998).

vary significantly. Therefore, measurements of water uptake are required at different sizes representative of the different modes and at different relative humidities between 10% and 95%. For ground-based platforms, these measurements are required at approximately 30 minute time resolution, whereas for aircraft measurements a time resolution of one-to-five minutes or less will be necessary.

Size-resolved water uptake measurements must be performed simultaneously with size-resolved particle chemical composition measurements so that different compositions and sources of particles can be related to the observed hygroscopic properties. Techniques that can delineate the influences of particle surface composition from bulk compositional effects are needed so that the role of gas-to-particle conversion processes can be separated from the role that different sources of directly emitted particles might play in determining particle hygroscopicity.

Recently, particle surface characteristics and particle morphology have been implicated as important factors in determining how particle and labile gas-phase species interact. New measurement techniques are required to elucidate particle non-sphericity and relevant surface characteristics so these properties

Measurements of gas-phase precursors such as sulfuric acid, ammonia, and organics are critical to improved understanding of new particle formation in the atmosphere.

There is an important need for instruments to identify the presence of newly formed particles and to quantify the rate of formation and growth of new particles.

may be related to processes that alter the ambient aerosol.

An important area of instrument development relevant to processes that alter the ambient aerosol is fast-response aerosol instruments for quantifying the effects of meteorological and other processes on the ambient number and chemical size distribution. For example, simultaneous size-resolved particle water uptake and ambient relative humidity measurements on-board aircraft would allow better estimates to be made of boundary layer light scattering properties.

There are numerous scientific questions relating to aerosol-cloud interactions that can be addressed through new instrument development. Techniques such as eddy correlation could be employed to better understand entrainment and deposition processes. Residual and other layers mix with the growing daytime boundary layer, and this mixing can influence the chemical and physical properties of the ambient aerosol. Previous work (Russell et al. 1998) has shown that even in remote marine regions, complex bidirectional mixing of gases and particles occurs across different intermediate layers in the planetary boundary layer. The mixing processes over land surfaces will be even more challenging to quantify due to strong diurnal forcing and associated atmospheric dynamics.

To complement ground level and in-situ aircraft measurements and place them in a larger context, continued development and evaluation of remote sensing instrumentation to measure aerosol properties as a function of altitude is required, including ground and airborne lidar for aerosol vertical and horizontal structure (Figures 5.8 and 5.9). For

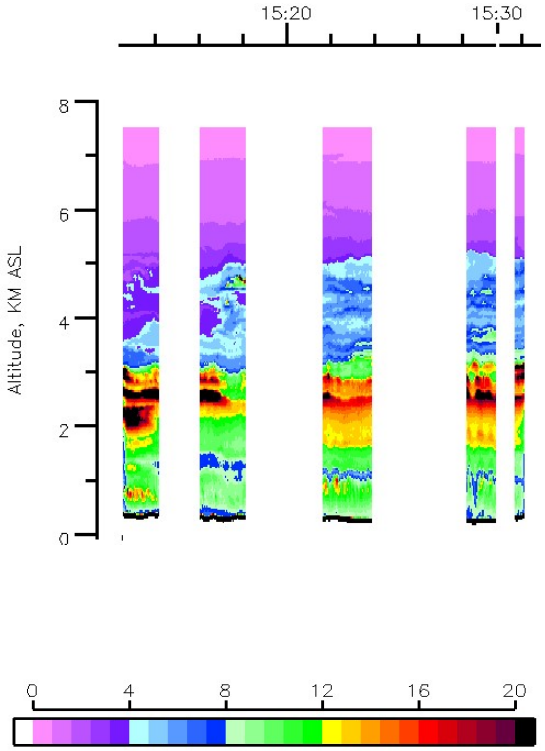


Figure 5.8 Airborne Lidar image of vertical and horizontal structure of aerosol extinction above ARM SGP site in north Central Oklahoma on 11 July, 1997. Color bar gives relative aerosol extinction shown as a function of altitude (left ordinate) and time (UTC, top abscissa) with latitude (˚N) and longitude (˚W) shown on lower abscissa. From Browell (1999).

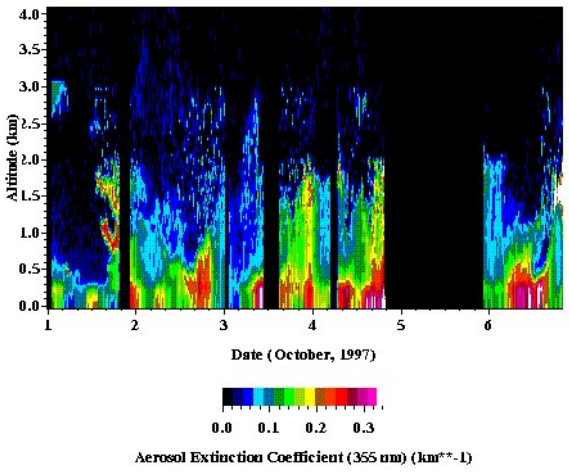


Figure 5.9. Time-height profile of aerosol extinction coefficient as measured with Raman Lidar at the ARM Southern Great Plains site in north central Oklahoma over 7 days in October, 1997. Note substantial variation in loading and vertical structure of aerosol in this period. From Ferrare (1998).

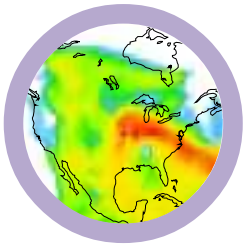
example, pulse compression millimeter radars are sensitive enough to measure cloud boundaries and gain great scientific power when used in conjunction with lidar. Increased detection sensitivity and range/temporal resolution is required in future lidar instruments. Co-located sonic radar systems could provide important meteorological data to elucidate how boundary-layer processes such as entrainment and cloud processing alter aerosol properties.

Finally it should be noted that field measurements of aerosols, as well as some laboratory studies, require sampling inlets. The ideal inlet and sampling system efficiently transports the sample without losses or modification to the observing instrument or instruments independent of the sampling platform. However, state-of-the-art inlets are far from ideal, particularly for aircraft applications. Sampling uncertainties related to volatilization or condensation of water vapor, organic vapors and others volatile species, flow distortion and related aspiration inefficiencies, and other particle loss mechanisms are often far larger than the uncertainties of the sampling instruments themselves, and despite much work these errors remain poorly constrained. The problem of artifact particles due to droplet shattering inside aerosol inlets during in-cloud and in-haze sampling must be resolved so that the influence of clouds and elevated relative humidity environments on ambient aerosol properties can be properly investigated.

Science Support Requirements

It is expected that TAP principal investigators who participate in Instrumentation Development and Advanced Characterization activities will take part in TAP Field Campaigns by conducting measurements at the supersite or on the research aircraft. In either case it will be required that the TAP Science Support Team provide space, power, and communications resources. TAP Science Support will ingest data into the TAP data system and facilitate exchange of data between investigators and provide external data as required during Field Campaigns and subsequent analysis and interpretation of results. TAP Science Support will provide appropriate archiving of data from IDAC investigators.

5.3 Modeling



Important as measurements are to understanding tropospheric aerosols, merely making measurements and reporting them will not achieve the objectives of TAP. The portfolio of results need to be synthesized and integrated. This generally is done with the assistance of complex mathematical models that describe the air quality conditions as a function of the forcing of meteorological properties, emissions patterns, air chemistry and deposition over the spatial and temporal regime of concern. Most air quality models of this kind have been focused heavily on conditions that represent the regulatory bounds for averaging and for outputs representing ambient concentrations. However, considerable effort is needed to create a new hierarchy of models that fully integrate gas-phase and condensed-phase processes, including accounting for cloud processes, with emissions and meteorological features in the spatial regime to be investigated. Such models, which predict ambient concentrations by representing the processes that take place between emission and receptor are referred to as "source models" to distinguish them from models which are based on empirical relations between ambient concentrations and emissions, known as "receptor models" (Seigneur et al., 1999).

A hierarchy of models is needed to fully integrate gas-phase and condensed-phase processes, including cloud processes, with emissions and the controlling meteorology.

The new codes will require bookkeeping on a very large number of variables, and will demand a computational scale similar to those used in today's climate models. The models themselves can create outputs of a wide range of variables, and can be used to investigate phenomena over a variety of time and spatial resolution that are important for understanding atmospheric processes. The models to be developed and adopted for TAP will be flexible in their access to output and their use of input data even though their form may be far more complex than current

regulatory models. The aim of these models is to provide a means not only for analyzing, inter-comparing, and interpreting the experimental results, but also ultimately for extending and qualifying models used for air quality management practices.

The modeling component of TAP takes its direction from the program's emphasis on obtaining a process level understanding of the life cycle of tropospheric aerosols. A detailed description of particle nucleation and growth is best obtained by studying these process in isolation. This requires a zero dimensional (box) model. However, in order to study regional scale implications, the detailed process level description has to be combined with all of the complications of the 3 dimensional world, including flow fields, boundary conditions, emissions, and mixing of air masses. This requires a regional scale chemistry - transport model. It is proposed that TAP concentrate its efforts on 1) developing advanced zero dimensional descriptions of aerosol chemistry and microphysics leading to computer codes (modules) that can be exercised either alone or within a regional transport model and 2) carrying out regional scale simulations using these modules incorporated in existing transport codes. While recognizing the desirability of having many independent approaches to doing the transport calculation, it is also recognized that there are efficiencies in having a common framework. It is therefore proposed that TAP make available a community transport model that can be used by investigators to inter-compare aerosol modules, explore the regional scale consequences of aerosol interactions, and analyze field observations.

The modeling component of TAP takes its direction from the program's emphasis on obtaining a process level understanding of the life cycle of tropospheric aerosols.

"Cubic meter" models

A detailed description of aerosol nucleation and growth involves the processes of gas phase chemistry, homogeneous and heterogeneous nucleation, transfer of material between the gas and particulate phase, and particle - particle interactions. Some of these processes are described by kinetic relations whereas others are best described in terms of phase equilibria. Different methods are employed by different investigators to represent these processes, and evaluating the situations that favor one or another methods is a matter of active research. For example, aerosol size distributions can be represented in models by discrete size bins, modes of specified or evolving mean radius and width, or moments of the size distribution. For many problems the governing kinetic rate expressions and thermodynamic properties have not yet been quantified and in some cases have not been even identified.

The field, laboratory, and theoretical components of TAP are expected to provide the information needed to construct more accurate detailed models. For example, in order to predict new particle nucleation rates under the range of conditions expected in the field programs, current theories will have to be extended to include ternary mixtures, organics, ion induced process, and the effects of near-cloud environments. Accurate descriptions of aerosol growth processes will require laboratory studies on the thermodynamic properties of complex mixtures (including organics), accommodation coefficients, and rate constants. Laboratory data is also needed to address the possible effects of aerosol mediated reactions on gas phase chemistry. Based on suggestive but limited evidence, these reactions can constitute a sink for free radicals and a source of NO_x and reactive Cl. Predictions of the radiative effects of aerosols will require laboratory results and a theoretical framework for determining the optical properties (including long wave interactions) of organics, carbon containing and mixed particles.

Field investigations are needed to provide the ambient conditions which control the chemical and physical aerosol interactions and to test whether models are representing processes correctly. For some aspects of aerosol evolution, it is the absence of field data which is the limiting factor in constructing an accurate model. For example, the fundamental physical and chemical relations describing in-cloud oxidation are known moderately well but there is difficulty in predicting the end results on the atmosphere. Cloud chemistry data and aerosol size spectra before and after cloud processing are needed.

Box models will also be used in the analysis of field observations, in particular for describing the rapid chemistry and microphysics that is in a near equilibrium with the observed mixture of long lived species (the contents of a small volume of air, say a cubic meter). This type of calculation yields rate information that cannot be obtained directly from observations, for example the rate of new particle formation. This type of calculation also yields the concentrations of transient species such as free radicals and H_2SO_4. Thus, the concentration of H_2SO_4 can be calculated from a model that uses observed concentrations of trace gases to determine the oxidation rate of SO_2 and the observed size distribution of aerosol particles to determine the surface loss rate of H_2SO_4. The model can be tested in a "closure" experiment by comparing calculated and measured values for H_2SO_4.

Box models can also be used to follow the time evolution of an air parcel without the complications that occur in the real 3 dimensional world. Such calculations might be used to study the changes in aerosol composition and size distribution in a plume that is advected away from a source region.

The challenge facing TAP is not only to improve the theoretical basis for describing atmospheric aerosols, but also to arrive at a compact description that can be used in regional scale transport models. Operationally, this will require using detailed box models to explore aspects of the chemistry and physics of aerosols; then, by way of systematic sensitivity calculations, construct an abbreviated description that can be used to convert the detailed box model into a module that is both accurate and capable practical implementation within in a transport code.

Regional- and larger-scale Modeling

Because the real atmosphere is three-dimensional, and because air parcels never exist in complete isolation, it is necessary to embed the aerosol calculations within a larger scale transport model. This model should be capable of resolving the spatial distribution of pollutants within the 200 by 200 km field observation area. Regional calculations will yield process level information on chemical pathways, yields, and rates; concentration fields that can be compared with observations; and estimates of the imports into the experimental region and the exports from the region.

Transport models will be used to develop an experimental plan in preparation for a field campaign. It is particularly important to determine whether an anticipated signal due to a process under investigation has a large enough amplitude to stand out over the noise of all other confounding variables. After the field experiment, transport models (and cubic meter models) will be a major part of data analysis. It is these requirements that lead to a modeling focus on the same scale as the experimental arena. A very important component of the modeling activity must be the characterization of the meteorological variables that are responsible for aerosol transport and the representation of this transport in the model.

Because the real atmosphere is three-dimensional, and because air parcels never exist in complete isolation, it is necessary to embed the aerosol calculations within a larger scale transport model.

The geographic domain of the transport model is expected to be larger than the 200 by 200 km experimental arena, albeit with coarser resolution outside of the field observation area. A larger domain decreases the dependence of model solutions on boundary conditions and allows for the calculation of transport to and from a regional size area that will at times be part of a similarly polluted air mass. A regional scale context is also important as that is the scale upon which many energy related policies are evaluated. Models developed under TAP will provide powerful tools for the regulatory and health effects communities to use for determining source - receptor relations, exploring the effectiveness of various energy policies and technologies in reducing PM 2.5 levels, and for determining exposure due to specific size and chemical components of the ambient aerosol mixture. The global change community will find the TAP transport models useful in yielding a detailed description of near-source regions.

It is proposed that a community model be made available for doing regional scale

calculations. One possibility is EPA's Models-3/Community Multiscale Air Quality (CMAQ, 2001), which provides a flexible framework for incorporating advanced modules describing aerosols and gas-aerosol interactions. It is anticipated that most of the model development projects under TAP will be directed at the smaller box model scale and will yield products that can be used within the community model, or alternatively within other existing transport codes. As part of the support for a community model, the TAP Science Support Team will provide the needed input files such as wind fields and emission inventories.

Computational requirements for the transport model will be extremely large. Fast parallel computers will be needed. It is important that TAP establish links with computational efforts being pursued in DOE.

An important component of the development of highly sophisticated mathematical models is an evaluation procedure. Protocols are needed for evaluating the representation of individual processes within the models, for determining the ability of the models to predict quantities such as source-receptor relations which cannot be directly measured, and also for determining the overall ability of the models to predict concentration fields. The protocols needed for these different applications are by no means obvious and will have to be determined as part of the TAP research effort. Having a component of the evaluation procedure that focuses on process representation is motivated by the experiences of the photochemical modeling community. They have found that a good comparison between calculated and observed concentrations can be fortuitous and does not necessarily imply that the model has correct chemistry, a feature that could lead to erroneous predictions of the effects of emission controls. Conversely a poor comparison with observed concentrations may be due to errors in emissions and/or wind fields, and does not necessarily imply that the chemistry or physics of the model is incorrect. Process analysis will examine the changes in the amount of substances in each model grid cell due to the various processes of concern--emissions, advection, diffusion, deposition, phase changes, photochemistry, coagulation, dissolution--and compare these to measurements and to physical-based understanding. Process tests will include such items as closure experiments for OH, H_2SO_4, and light scattering coefficient, σ_{sp}; comparisons between observed and calculated concentration ratios near source regions for testing emission inventories; use of tracers such as ^{14}C to evaluate the calculated contributions to aerosol mass from biogenic compounds; and using ratios of substance that react at different rates to test the oxidation chemistry.

Field programs have to be designed with these tests in mind. These tests, however, do not completely take the place of an overall evaluation in which observed and calculated concentration fields are compared. There are many ways in which to do such a comparison and a task will be to arrive at a protocol that is relatively robust with respect to the stochastic components of the driving meteorological fields. Another task will be to conduct comparisons between different modeling groups, perhaps involving a community model run with different versions of an aerosol module. These types of comparisons are often an excellent way to evaluate the strengths and weakness of various approaches.

Interaction between modelers and laboratory and field investigators is envisioned as a two-way street in which the modelers provide questions that need to be addressed by the laboratory and field measurements and the measurements provide input and insight for the modelers.

Large Scale Computation

Computational requirements for the transport model will be extremely large. The number of computed variables will be larger than in a photochemical model of comparable sophistication because of the need to resolve the aerosol distribution by both size and composition. It may be necessary also to resolve the distribution into different morphology classes. Spatial resolution requirements are also stringent because of the need to resolve features within the 200 by 200 km experimental arena. Fast parallel computers will be needed. It is important that TAP establish links with computational efforts being pursued in DOE.

Science Support Requirements

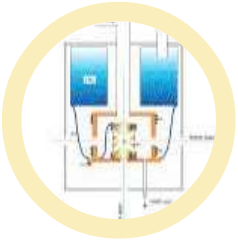
TAP Science Support will be responsible for acquiring meteorological data and for preparing meteorological fields for use by modelers. TAP Science Support will acquire satellite data; air quality data and emission inventories and make these data available to TAP investigators and the broader scientific community to facilitate process modeling and comparison of models and observations. This activity will make use of modern Internet and web-based technologies to enhance collaboration among TAP investigators at multiple institutions and hasten reaching TAP goals.

5.4 Laboratory Studies and Theory

This component of TAP is designed to generate the fundamental chemical and physical knowledge that is essential for understanding the processes (formation, transformation, and deposition) that control the life cycle of tropospheric aerosol particles. Only with a fundamental understanding of these complex phenomena can the predictive modeling tools that represent the final product of TAP be fully realized. This element will be tightly coupled to the field, modeling and health community activities needs, deriving its research subjects on the basis of public health studies and field observations, and generating the appropriate data to fulfill the most critical modeling needs.

Although field observations are the key to defining the *state* of a system, they are inherently limited in their ability to characterize *processes* (*e.g.,* identify specific reactions and determine their rates). To develop the appropriate theoretical framework, to test hypotheses, and to produce the appropriate inputs for the models requires going beyond observation into experimentation under controlled conditions. Experimental and theoretical studies will aim to characterize the fundamental properties and processes that control the formation, growth, evaporation, chemistry, phase transformations, optical properties and dry deposition of atmospheric aerosols.

Recent advances in single particle analysis techniques have shown that tropospheric aerosols are composed of a mixture of a large number of substances such as inorganic salts and acids, soot, mineral dust, semi-volatile organic compounds, transition metals and others. Until very recently field observations of aerosol compositions were incapable of providing information on the chemical composition of individual particles thereby distinguishing between internally and externally mixed aerosols. In contrast laboratory studies have for the most part focused on single component model aerosols.



It is crucial that experiments clearly define their relevancy to the pressing need to predict atmospheric aerosol processes. As an example, lab experimental conditions should span realistic environmental conditions (temperature, partial pressure, relative humidity, and particle sizes). Past research has shown that unrealistic environmental conditions such as a high partial pressure of a reactive component can place a system into a different and unrealistic physical regime. The laboratory and theory work sponsored by TAP will focus on aerosol systems of sufficient complexity to address questions raised by the field and modeling efforts.

Laboratory and theoretical studies are essential to understand the processes that control the life cycle of tropospheric aerosols.

The response of field instruments to the complex aerosols in the ambient atmosphere is far from understood. Required are laboratory experiments to provide essential "calibration" of the field instruments. Therefore proposed research projects that focus on the calibration of field instruments using characterized aerosols within the controlled environment of a laboratory are needed. Other examples of proposed research projects that would fall into this category would be collaborations between a laboratory group and a field group to investigate a particular physical property such as the real refractive index of plausible aerosol compositions to explain observations made by an optical instrument in the field.

The following describe key research issues that need to be resolved to insure a successful fulfillment of the goals of TAP.

Areas of Research

New particle formation

Atmospheric aerosols can be formed directly by primary emissions of particulate matter or secondarily through gas-to-particle conversion processes through homogeneous nucleation. The freshly nucleated nanometer sized particles can eventually grow through the condensation of gas-phase molecules and coagulation with other existing aerosol particles.

In the real atmosphere nucleation and particle growth are always in competition. Being able to represent particle formation is central to understanding aerosol evolution and representing it in models.

The importance of the nucleation process to the overall atmospheric aerosol loading and its detailed mechanism remains one of the key unknowns in atmospheric aerosol research. For example, the recent development of new field instrumentation made it possible to quantify atmospheric nucleation events in various locations and under a great number of conditions. When these field observations were compared with predictions from classical nucleation theory (employing the commonly assumed nucleation mechanism), a discrepancy of many orders of magnitudes was uncovered. The field data indicate that, except for in the very remote atmosphere, the true rate of new particle formation is much higher than that predicted by the prevailing binary classical nucleation theory. Several hypotheses have been suggested to explain these observations most of which invoke a ternary nucleation process, such as ammonia or organic compounds added to the common binary system of water and sulfuric acid.

To complicate the issue, current nucleation theory largely relies on bulk thermodynamic properties to describe the chemical potential of the critical cluster that may range from several to tens of molecules. It would highly desirable to develop molecular models that will make it possible to predict the rates from first principles. The theoretical approaches are at a promising state, but critically require benchmark experiments. Experimental studies of nucleation rates versus concentration of condensing molecules in the gas-phase, where the contribution from contaminant species is eliminated, are required for multicomponent systems. With such experimental studies and improved homogeneous nucleation theories, the task will be to determine the atmospheric conditions under which homogeneous nucleation processes play a significant role in aerosol loading and the conditions under which homogeneous nucleation can be ignored.

Two important types of secondary aerosols are sulfates and organic compounds. Field and modeling studies have shown a clear correlation between sulfate aerosol production in the troposphere and the presence of volatile sulfur compounds, such as dimethyl sulfide (DMS) and sulfur dioxide (SO_2), as well as the direct precursors to aerosol formation such as H_2SO_4. The mechanisms for formation of organic aerosols are much more complicated because of the complex mechanisms for oxidation of the volatile organic compounds (such as isoprene and terpenes) to form the secondary products (aldehydes, ketones, and organic acids) that can condense as aerosols. Laboratory and theoretical studies are needed to understand the mechanisms and kinetics of the gas-phase oxidation of volatile organic and inorganic species to form the gas-phase precursors to nucleation of new aerosol particles. It is only recently that first principles calculations of reaction rates and thermodynamics and intermolecular potentials have become accurate enough to fill out regions of parameter space not filled by experimental data.

In the real atmosphere nucleation and particle growth are always in competition. The relative rates of these two competing processes will determine the influence that nucleation imparts on the size and chemical distributions as well as the total mass loading of the aerosol.

The nucleation events described above tend to produce a large number of freshly nucleated particles. There might be a second mechanism that continuously produces a low-level number of particles.

Being able to represent particle formation in models is clearly central to any aerosol program. Only through careful experimentation can the various hypotheses be tested. TAP will conduct laboratory and theoretical studies directed towards measuring nucleation rates and elucidating the mechanisms that lead to the observed rapid nucleation rates.

Aerosol condensation and growth kinetics

Rates of condensation and evaporation (volatilization) of gas species onto and from aerosol surfaces are fundamental to most aerosol microphysical processes. Current experimental and theoretical studies of these processes have been limited to relatively simple chemical systems, either pure water or stoichiometric inorganic systems (e.g. sulfates and nitrates). In view of the chemical complexity of atmospheric aerosols that is increasingly apparent, laboratory studies of gas uptake rates need to investigate appropriate mixtures of inorganic and organic species. The dependence of condensation/ evaporation processes on heterogeneity of bulk and surface composition is largely unknown. This will require the development of multiple experimental techniques that can generate and monitor the evolution of particles with complex chemical composition.

In addition, there is currently no theory that can comprehensively relate molecular scale interactions to bulk scale experimental observations. Detailed understanding of gas/surface interactions over the full range of atmospheric particle size (≤5 nm to ≥1 μm) is required in order to accurately model aerosol microphysics, both in interpreting atmospheric observations and ultimately for predicting atmospheric aerosol processes.

A case in point is the ternary $NH_3/H_2SO_4/H_2O$ system that which largely represents the aerosol nucleation/growth process in conventional aerosol models. Even for this relatively simple system, gas uptake rates have not been measured over the full range of compositions. The direct and indirect effect of other species such as organic compounds on aerosol nucleation and growth has not been investigated. Gas uptake measurements need to be extended to a wide range of particle parameters, including variable temperature, chemical composition and size. The effects of co-deposition of multiple species on surface properties and gas/surface kinetics will be critical to understanding realistic complex systems. Because of these dependencies, it is important for experimental and theoretical studies to closely model the environmental conditions observed in the field. For example, a growing ammonium nitrate particle might have a labile adsorbed monolayer of organic oils that controls its growth and evaporation. The growth of such an ammonium nitrate aerosol might depend strongly on the local mix of gas-phase semi-volatile organic oils, and differ greatly from a pure laboratory-synthesized ammonium nitrate aerosol.
The TAP research program will derive the appropriate accommodation coefficients that are necessary to describe this phase of a particle's life cycle, and will emphasize the coupling of laboratory, theory and field programs to develop the detailed framework for model parameterizations of gas/particle interactions in the atmosphere.

There is currently no theory that can relate molecular-scale interactions to bulk-scale experimental observations.

Relative humidity and phase transformations

A large fraction of atmospheric aerosols contain hygroscopic substances that interact strongly with water vapor. Particle size, composition, optical properties and phase are all functions of the ambient relative humidity. The phase of the aerosol is important because the properties of wet and dry particles differ substantially. As compared to dry particles, wet particles tend to be larger, have lower refractive indices in the visible, higher absorption in the infrared, and have larger reaction probabilities for several important chemical reactions. Their fate when inhaled is also strongly dependent on their hygroscopicity.

An aerosol composed of a single hygroscopic substance will undergo a sharp solid to liquid phase transformation when the relative humidity reaches a characteristic deliquescence point. Further increases in relative humidity results in particle growth and the formation of a dilute solution droplet. At relative humidity conditions slightly above 100% the same particle can be activated to form cloud droplets. When the relative humidity is then decreased the droplet reversibly shrinks in size, except that it does not transform back to the solid phase at the deliquescence point but instead becomes supersaturated and efflorescence

Each gas-phase molecule collides with an aerosol particle approximately once per minute. It can become adsorbed, react, or bounce back with no consequence. Reactions between gas-phase species and aerosol particles can directly influence the composition, phase, and morphology of atmospheric aerosols and gas-phase composition.

(crystallization) occurs at a much lower relative humidity. The behavior of particles with multiple components will obviously be much more complex. At present, data for such deliquescence and efflorescence cycles are available for only a few model systems, and only over a narrow temperature range.
Phase transformations can also be a strong function of particle size. This is particularly true for efflorescence, which is kinetically controlled. As a rule smaller particles effloresce at lower RH. What that dependence is and how that rule is affected by the presence of other components such as insoluble mineral dust remain open questions. Even the thermodynamically controlled deliquescence point uptake curves become size dependent for particles smaller than 20 nm.

TAP will conduct experiments on aerosols of realistic compositions aimed at developing the database and the theoretical framework to be able to simulate the effect of RH on particle size, composition, and phase.

Heterogeneous chemistry

Each gas phase molecule collides with an aerosol approximately once per minute. It can become adsorbed, react, or bounce back with no consequence. Reactions that occur between gas-phase species and an aerosol particle, called heterogeneous reactions, can directly influence the chemical composition, phase, and morphology of atmospheric aerosol particles. The same reactions will clearly also modify the gas phase composition.

Reactions on aerosol particles are inherently complicated. Not only are the aerosols mixtures of many components, but the surface properties of the aerosol may be vastly different than the bulk properties. For example, even in very dry air a monolayer of tightly bound water may exist on an atmospheric aerosol surface. Such a surface may provide an aqueous-like environment for chemical reactions such as sulfur oxidation. As a rule surface phases can have very different thermodynamics (and kinetics) than bulk phases. For this reason, it is crucial to employ analytical tools that can probe the outer-most layers of an aerosol, and identify the true reactive phases and sites.

In addition to quantifying the effect of heterogeneous processes on the aerosol phase, it is desirable to understand how the presence of aerosols influences gas phase processes through various mechanisms. For example, during the night, a significant fraction (20 to 60%) of NO_x may be processed by wet aerosols through the hydrolysis reaction of N_2O_5 to produce HNO_3. Yet, the effects of possible organic compounds on the surface of such wet aerosols on the hydrolysis reaction rate of N_2O_5 are presently unknown.

Current tropospheric models overpredict the ratio of nitric acid to NO_x compared with field observations. Recent experimental and theoretical results suggest that nitric acid may be heterogeneously lost on ice, mineral aerosol, or soot particles. Other studies suggest a role for heterogeneous formation of HONO, a molecule that may be responsible for the early morning production of ozone. However, the role of aerosols in controlling NO_y/NO_x chemistry is still uncertain. Experiments under realistic atmospheric conditions of temperature, relative humidity, and gas phase partial pressure, as well as theoretical studies are necessary to answer these questions.

Aerosols may also affect HO_x chemistry by providing sites where radicals like OH and HO_2 are deactivated. While the importance of OH in gas phase oxidation pathways is well established, the possible reactions of OH with condensed organic compounds is unknown. However, such reactions have the potential to functionalize the surface of the aerosols as they age. This would impact the chemical reactivity of the aerosols, their hygroscopicity, and their optical properties. Furthermore, the reaction of OH with organics, to form alcohols for example, will change their reactivity and miscibility.

In addition to the above examples, it has also been hypothesized that active chlorine can be released from sea salt aerosols through an unknown mechanism.

Quantifying the effects of aerosols on gas-phase concentrations is an integral part of TAP. TAP research projects will identify the key heterogeneous reactions and quantify their effects both on the aerosol particle characteristics and the gas-phase concentrations of certain chemical species.

Optical properties of tropospheric aerosols

Several of the most important effects of tropospheric aerosol particles, visibility reduction and climate forcing, are directly related to the ability of the particles to scatter and absorb solar radiation. In addition, a majority of aerosol field instrumentation and all remote sensing techniques rely on deriving aerosol physical parameters using known and assumed optical properties of atmospheric aerosols. To date the experimental data covering the optical properties of complex aerosol particles is scarce and the theoretical framework presently employed for extrapolating these data to more complicated cases has not been well tested. In conjunction with the field program the TAP will derive the optical data that are required to invert field observation into aerosol loading.

Organic species in atmospheric aerosol particles

The term organics refers broadly to the class of a large number of compounds composed of carbon, hydrogen, nitrogen, sulfur, phosphorus, and oxygen that may include soot, aldehydes, ketones, organic acids and surfactants. The very fact that they are classified so broadly reflects current lack of knowledge. Recent field data indicate that these compounds are ubiquitous to the surfaces of aerosols, even those nominally inorganic in bulk composition. Many of the organics are derived from the degradation products of naturally emitted VOCs such as isoprene, while others are of anthropogenic origin. The role of organics in the processes that control the life cycle of tropospheric aerosol particles and their effects on health, visibility, and climate, is a key topic that will be addressed by the Laboratory and Theory component of TAP.

There currently exists a lack of knowledge concerning the type and concentration of organic compounds within tropospheric aerosol particles. In order to assess the role of organics on the life cycle of aerosols it is critical that the experimental theoretical and field program studies work closely together with the field component in order to identify the most important compounds for investigation.

Aerosol transport and deposition processes

The focus of the TAP mission is to characterize tropospheric aerosols in relation to health hazards and visibility reduction caused by urban pollution centers. The spatial limits chosen are on the order of 200 km from a source, which limit the aerosol processes investigated to those that occur within such a near field. While some particles will be transported beyond these limits, several important aerosol loss processes will occur within these limits and are important for understanding the spatial and temporal aerosol mass loading from pollution plumes. Two such important processes are dry deposition rates and the scavenging of aerosol particles by larger cloud droplets. An aspect of this research program will be the quantification of the rates of aerosol loss through these mechanisms.

The term "organics" refers broadly to the class of a large number of compounds composed of carbon, hydrogen, nitrogen, sulfur, phosphorus, and oxygen that may include soot, aldehydes, ketones, organic acids and surfactants. The very fact that they are classified so broadly reflects current lack of knowledge.

Science Support Requirements

1. Providing Standards and Facilitating Comparisons of Aerosol Sources

In order to facilitate intercomparisons between laboratory experiments, field instrument calibrations, and field observations it will be important to develop and characterize standard aerosol generation methods that can be used by experimentalists and field scientists.

2. Providing Sources of tropospheric aerosols for lab research

Methods for the collection of real ambient tropospheric aerosol particles from the field to be transported back to the lab for experimental measurements of their reactivity, growth, morphology, etc. will be pursued. These will allow both access to instrumentation that is not field-portable, and will help bridge the gap between lab-created aerosols and real-world complex ones.

3. Data Bases

TAP is inherently multi-investigator and multi-disciplinary in nature. Thus, it is important to provide for the rapid and effective communication of results between all elements of the TAP team. The development of data bases and other means for sharing results in common formats will be an important attribute of TAP.

4. Facilitating Collaborative Use of Specialized Laboratory Instrumentation and Theoretical Models.

Progress in the study of complex materials and processes characteristic of atmospheric aerosols will require substantial and ongoing advances in experimental and theoretical capabilities. This will require a significant investment of TAP resources. To optimize the impact of this investment, every effort will be made by resource developers to share these resources (through collaborations and other means) as widely as possible with other TAP researchers. TAP Science Support resources will be crucial to facilitating this collaborative effort.

6. Science Support Implementation

The Science Support component of TAP is seen as crucial to maximizing the productivity of the TAP Science team and thereby helping to achieve the objectives of TAP. The responsibilities of TAP Science Support are delineated into the following components:

Support for Field Studies. Support necessary to conduct TAP Field Campaigns. Campaign planning and logistics; conducting baseline measurements at TAP supersite and satellite sites and on aircraft platforms, aircraft support and coordination, QA/QC. Maintaining a web-based campaign logistics and data server.

Data system and archive. Incorporate measurements into data base/archive to facilitate model development and testing. Make data available to TAP investigators and investigators from cooperating agencies. Acquiring external data required by TAP investigators, including meteorological and satellite data.

Support for modeling activities. Preparation of meteorological drivers and gridded data sets for comparisons; establishing and maintaining community models supporting swap in/out of modules.

Facilitating scientific communication among TAP investigators and serving the needs of the interagency aerosol research community and the broader scientific community. This activity will include maintaining a data base of publications, organizing special sessions at conferences, special issues

While ultimately the Science Support activities of TAP will depend on the requirements of the Science Team, many of the required activities can be reasonably anticipated for planning purposes. The activities outlined here are based on past experience in large-scale Field Campaigns conducted by DOE and other agencies (NOAA, NASA, NSF), often involving multi-agency coordination.

6.1 Support for Field Studies

Support necessary to conduct TAP Field Campaigns as outlined in each Field Campaign Science Plan will be provided by TAP Science Support. This effort will include preparation of an Operations Plan, providing the aircraft- and ground-based platforms and baseline measurements for each Field Campaign, conducting baseline atmospheric chemical, physical, microphysical, and meteorological measurements, and providing the data to the Science Team investigators and to the TAP archive. TAP Science Support will also assist Science Team Investigators subsequent to the Campaign in analysis, interpretation, and modeling.

The Operations Plan for a given campaign, will provide for carrying out the measurements specified the Science Plan consistent with available technology and resources. To achieve this, TAP will need to develop a management structure to accomplish the detailed planning that is needed to conduct the large scale Field Campaigns that are planned for TAP, and to operate the required facilities in the field. A proposed management structure is shown in Figure 6.1. Planning and implementation activities will be overseen by the Chief of Operations. Each function in Figure 1 will be accomplished by a team of scientists, technicians, and other support personnel. Examples of the tasks for each of these teams are given in Table 6.1.

While ultimately the Science Support activities of TAP will depend on the requirements of the Science Team, many of these activities can be reasonably anticipated for planning purposes.

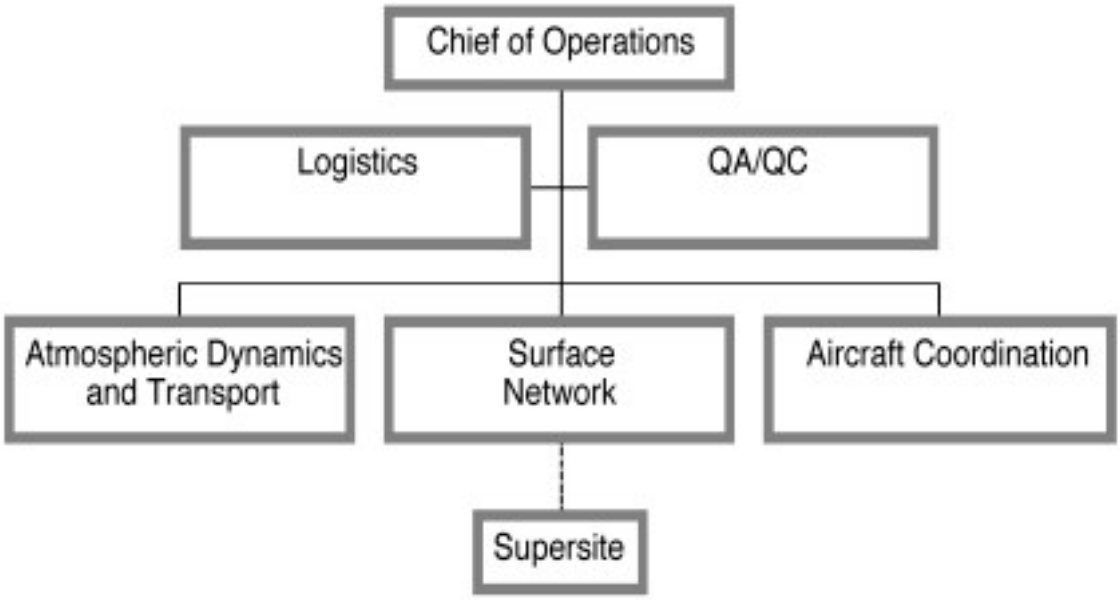


Figure 6.1 Organizational structure for TAP field support activities

The measurements to be made by TAP Science Support and the instruments to be employed will be selected under the guidance and direction of the TAP Field Investigators Team. TAP Science Support will be responsible to set up, and calibrate the baseline instruments prior to the campaign, operate these instruments during the campaign, and provide quality assured data to the data archive upon completion of each campaign.

TAP Science Support will be responsible for the baseline measurements at the surface sites, and on at least one aircraft platform. Baseline measurements are defined as those measurements that utilize tested and established techniques. The need for specialized one of-a-kind principal investigator instruments is also recognized. These will be funded either through the normal peer-review process, or by sub-contract. For the surface sites, the measurements that TAP will be responsible for are listed in Table 6.2 as are the desired specialized measurements. TAP expects to deploy one supersite and up to six satellite sites for each Field Campaign. As indicated in Table 6.1 some of the baseline instruments will be provided for all sites, whereas others will be provided only for the supersite.

Table 6.1. Tasks of the Science Support Teams for operation of TAP Field Campaigns

Logistics	Quality Assurance/ Quality Control	Aircraft coordination
Support facilities for participating scientists Communications, computer networks Meeting rooms Laboratory space Housing Ground site infrastructure Access agreements Power Communication Trailer rental and siting Erection of sampling towers Safety plan	QA/QC Plan Management Standard operating procedures for routine monitoring Audits Performance Special studies (e.g., measurement inter-comparisons)	Assemble and install scientific equipment Maintain and operate baseline aircraft instrumentation Arrange aircraft logistics Hangar, lab, office space Interaction with FAA Coordinate Flight Plans Manage data Aircraft QA

Ground Based Network	Atmospheric Dynamics and Transport	
Procure and prepare instruments Locate and set up sites Operate instruments Calibrate, maintain, inter-compare Manage data	Identify and locate ground based instruments Profilers Lidars Sondes Towers Calibrate and maintain instruments Manage data	Provide Meteorological forecasting to support flight operations Daily briefings Trajectories High resolution models

Table 6.2. Measurements to be deployed at TAP surface sites

Aerosol, physical	Aerosol, chemical	Trace gases
Total Mass Coarse mode size distribution (2 – 10μm) Accumulation mode size distribution (0.08 – 2 μm) Aitken mode size distribution (0.02 – 0.08 μm)[1] Nucleation mode size distribution (0.003-0.02 μm) [1,2] Aerosol absorption Multi-wavelength light scattering Vertical and horizontal distribution (lidar)[1] Multi-wavelength Optical Thickness Aerosol hygroscopicity[1,2]	Bulk composition- To include EC, OC, principal inorganic anions and cations Size distributed composition (impactor) Single particle composition[1,2] Trace metals[1,2]	NO, NO_2, NO_y, SO_2, O_3, CO, Hydrocarbons H_2O_2[1], Aldehydes[1], HNO_3[1] OH[1,2], HO_2[1,2], H_2SO_4[1,2], NH_3[1,2] Perfluorocarbon tracers

1. To be measured at the supersite only.
2. Principal Investigator instrument to be funded through a Science Team proposal or by subcontract.

TAP Science Support will be responsible also for instrumenting and operating the principal aircraft used in the campaign. This could be the DOE G-1 aircraft, or an alternate aircraft of larger size and endurance, and it may vary from campaign to campaign. It is anticipated that the services of a remote sensing aircraft, and helicopter would be obtained by contract. Such aircraft are typically equipment with the instrument required for their respective tasks. Support is required to set up, and calibrate the baseline instruments, to operate these instruments in the field, and to provide quality assured data to an archive upon completion of each campaign. It is presumed that any aircraft TAP will use will be equipped with instruments to measure atmospheric state parameters, altitude, position, winds, and turbulence. A projected set of instrumentation that TAP Science Support will take the responsibility for providing is shown in Table 6.3

Table 6.3. Measurements to be deployed on in-situ aircraft measurement platform.

Aerosol, physical	Aerosol, chemical	Trace gases
Accumulation mode size distribution (0.08 – 2 μm) Aitken mode size distribution(0.02 – 0.08 μm) Nucleation mode size distribution (0.003-0.02 μm)[1] Aerosol absorption Multi-wavelength light scattering Aerosol optical depth[1]	Bulk composition- To include EC, OC, principal inorganic anions and cations Size distributed composition Single particle composition[1]	NO, NO_2, NO_y, SO_2, O_3, CO, Hydrocarbons H_2O_2, Aldehydes HNO_3 OH^1 , $HO_2{}^1$, $H_2SO_4{}^1$, $NH_3{}^1$ Perfluorocarbon tracers

1. Principal Investigator instrument to be funded through a Science Team proposal or by subcontract.

Significant support staff and resources will be required to maintain, deploy, operate this equipment in the field. It is anticipated that TAP Science Support will make use of existing capabilities within the DOE complex where available. Alternatively if a measurement is required and is otherwise not available, it will be the responsibility of TAP Science Support to acquire this capability by contract. A similar approach may be taken for chemical analyses of filter samples.

TAP Science Support will also have the responsibility of operating base line meteorological instruments. This would include at least three wind profilers, and one or more sites for the release of rawinsondes.

6.2 Data System and Archive

The TAP Data System and Archive activity is a key component of TAP Science Support. This activity will include a data management component to support fieldwork and modeling activities and to make TAP data available in a form suitable for archiving. The data management effort will be driven by the scientific needs of TAP. It will, as much as possible, use either public domain or commercial off-the shelf software and existing hardware and software resources. This data management effort will take advantage of the experience gained in other DOE efforts such as ARM and EMSL. Archiving will be coordinated with other air quality archiving under the auspices of NARSTO (NARSTO, 2000).

TAP Data Management (TDM) will be responsible for providing the communications infrastructure during campaigns that is necessary for communication among the participants and with the Internet. This support will include providing mobile modems via a cellular system and Internet service at the central command center.

Real-time satellite, radar and analysis products necessary for forecasting/nowcasting will be provided. At the TAP command center, a multi-user computing facility will be provided for use during debriefings for viewing data collected by participants and for performing

intercomparisons to aid in quality assurance efforts. TDM will work with TAP scientists to determine what auxiliary data will be needed to augment field measurements and will acquire these data. TDM will work with TAP scientists to specify the metadata that should be acquired to understand the context of the field measurements and will acquire these data. The TAP Web site will be used to communicate information about planned and on-going field campaigns.

TAP Modeling Support will make available meteorological data sets suitable for driving chemical transport and transformation models, thus facilitating model application and comparison.

TDM will work with TAP scientists to determine what data, in specified formats must be acquired to drive the TAP supported models and will provide these input data to TAP modelers via the TAP web server.

TDM will provide the capability of running TAP supported models via its web server. The modeler will be able to choose the input conditions, drive the model and examine intermediate and final products using visualization tools. By examining model runs at selected check points,, the modeler will be able to control and make changes to the current model run.

The TAP Web Server will provide timely information about the Program to the Science Support staff, the Science Team, DOE Headquarters and the general public. It will be used in the planning of campaigns, to communicate the status of ongoing campaigns and to distribute the data associated with the campaigns.

As discussed above, the TAP Web Server will run TAP supported models. It will also serve as the vehicle for advertising the contents of the TAP Data Archive and for data delivery.

The TAP data archive will serve as a repository for data and metadata collected in campaigns, the input required to drive TAP supported models and selected results of model runs. Data will be made available in standard formats via a Web based interface (NARSTO, 2000). Users of TAP data will be tracked so that they may be informed of updated versions of data they have received.

6.3 Support for Modeling Activities

Modeling will be a major component of TAP and a major client of TAP Science Support. It is intended that TAP Science Support facilitate the development, use, and evaluation of process models that represent atmospheric chemistry and aerosol microphysics. This can be achieved by acquiring and making available to TAP modelers (and external scientists) meteorological data sets suitable for driving chemical transport and transformation models such as forecast model output from the U. S. National Centers for Environmental Prediction (NCEP). These data serve as input to various Eulerian codes and can also be used to drive trajectory models.

The Modeling Support activity will also provide data sets of chemical and microphysical measurements that can be used for model evaluation. In this effort it will be necessary to acquire and make available appropriate external data, such as from state and local air pollution agencies. Likewise Modeling Support will provide gridded emissions inventories for use by modelers.

All of the results from TAP measurements must be integrated into a useful data set that can be used interchangeably for verification of models and interpretation of spatial and temporal variations. Visualization and analysis tools are required to examine this complex data set and elucidate the effects of boundary layer processes on the ambient aerosol. In designing new tools for observing particle chemical and physical properties and relevant meteorological processes, attention must be paid to how the data from each of the different instruments will be integrated so that a clear picture of the life cycle of ambient aerosols is created.

6.4 Scientific Communication

TAP will be a broadly distributed activity, with research taking place in some dozens of institutions. In such a situation there is a tendency for progress to be slowed by lack of ready means of communication among participants. To a great extent facilities for such communications exist, by email, file transfer, and the web, but those tend to be binary communications, or, in the case of the web, one-way broadcasts without exchange and feedback. It is intended in TAP to facilitate multipartite communications not just during Field Campaigns, but throughout the duration of the Program, to permit TAP investigators to engage in continuous scientific dialog. It is hoped that this approach will maintain the excitement and engagement of TAP researchers and lead to improved level of inter-laboratory collaboration and more rapid progress toward TAP objectives.

More traditionally TAP Science Support will maintain an electronic archive and data base of preprints and publications that will allow TAP investigators and others to readily download TAP publications so that TAP findings may be more readily and rapidly disseminated, again advancing TAP objectives. This is particularly important given the interagency interest in results of TAP research.

It is anticipated that increasingly scientific publication will take place by electronic publication, not just electronic images of static pages, but, for example, animations of model output and even executable models. TAP Science Support will endeavor to facilitate such novel methods of publication as an effective means of disseminating TAP results and make TAP a model for such advanced publication methods in atmospheric science.

Finally, TAP Science Support will assist in dissemination of TAP results through regular Science Team meetings, by organizing special sessions at national meetings of learned societies, and by arranging special journal issues dedicated to TAP findings.

TAP Science Support will facilitate novel methods of electronic publication as an effective means of disseminating TAP results.

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7. Deployment Schedule and Resource Requirements

The TAP Science Team will set the scientific goals and approach of the Program consistent with the Program Plan.

The principal operating elements of TAP will be the Science Team and the Science Support Team. Both elements are crucial to the success of TAP. These operating elements will be supported by a modest project support effort. The Science Team will set the scientific goals and approach of the Program consistent with the Program Plan. The Science Support Team will be responsible for providing operational resources and facilities to meet the requirements of the Science Team, mainly in support of Field Campaigns and Modeling activities.

The approach to meeting the requirements for both operating elements of TAP are outlined below. This Section presents the anticipated schedule of deployment of TAP activities in these two program elements and the corresponding resource requirements.

For a program of the complexity of TAP it is considered prudent to initiate the project gradually and allow it to reach steady-state level of effort over a few years. This approach will allow the Science Team to be filled by means of a succession of Announcements of Opportunity which can be tailored to the needs of the program taking into account previously awarded projects. Likewise such a staged initiation will allow the Science Support element of TAP to build up to meet the measurement and data acquisition requirements of the program over a period of time, as opposed to reaching full capability immediately following the onset of the Program.

For a program of the complexity of TAP it is prudent to initiate the project gradually and allow it to reach steady-state level of effort over a few years.

In order to accomplish the staged initiation of TAP it is intended that TAP conduct initial field projects in conjunction with major field projects being conducted by other programs. In this way TAP can benefit from the support facilities and ancillary measurements available from the agencies responsible for these programs and focus attention on measurements of aerosol precursors, aerosol properties, and aerosol evolution. Several important field projects and measurement activities that appear well suited candidates for TAP participation are expected to take place in the 2002 - 2003 time frame. These activities are briefly described here.

The DOE Atmospheric Chemistry Program is planning several large-scale field studies including a major study of atmospheric oxidants in the Northeastern states in summer 2003. TAP will benefit from the meteorological support and chemical characterization efforts available in these studies.

The DOE Atmospheric Radiation Measurement (ARM) Program maintains a highly instrumented site in north central Oklahoma at which the radiative influence of aerosols is measured on an ongoing basis and at which there is limited regular characterization of aerosol loading and properties. A TAP field study at this site could take good advantage of these ongoing measurements and historical data base to couple TAP chemical and microphysical measurements with ARM radiation and meteorological measurements. ARM would benefit from knowledge of the optical and microphysical properties of the aerosol, enhancing the interpretation of radiation measurements. TAP would benefit from the detailed vertical characterization of aerosol loading and relative humidity available from ARM remote sensing measurements.

The DOE Environmental Meteorology Program is planning a series of studies to examine boundary layer processes in stably stratified conditions, in conditions of weak or intermittent turbulence. Such studies will bring to bear state-of- the-art suite of wind profiling instrumentation and other instruments for characterizing flows and mixing (EMP, 1998),

thereby relieving TAP of the requirement to conduct these measurements and to translate them into flow fields. The EMP and TAP programs will be looking for opportunities to collaborate, including conducting joint field measurement campaigns at a location such as Salt Lake City. As the Salt Lake City basin often has a substantial aerosol loading, TAP might make a substantial early advance to its objectives by joining forces with this project.

The Environmental Protection Agency has established several "supersites" to characterize particulate matter, its constituents, precursors, co-pollutants, atmospheric transport, and source categories in several urban regions: Atlanta, Baltimore, Fresno, Houston, Los Angeles, New York, Pittsburgh, and St. Louis (EPA, 2000). The enhanced measurements at these sites would make these locations very attractive for initial TAP studies of aerosol processes and evolution.

The Air Resources Board of the State of California has been conducting a series of studies of particulate air pollution in Central California (CARB, 2000). A TAP field study in conjunction with such a field study would greatly benefit from the availability of key meteorological and emissions data from state and other agencies that would be complementary to TAP investigations into aerosol processes.

7.1. TAP Science Team

The Lead Scientist will be responsible for overall scientific leadership of the program and for representing the program within the scientific community. The Lead Scientist is expected to make a major intellectual contribution to the science conducted in the program, including analysis of findings and implications, writing technical papers, and fostering the aims of the program to conduct research that will achieve the objectives of the program, and to generate products that will meet the requirements of the air quality research and regulatory communities.

A key to the success of TAP will be its ability to attract outstanding investigators from within the DOE National Laboratories, including areas of these laboratories that have not been traditionally supported by ESD programs; from other Federal Agencies having expertise in atmospheric and aerosol science; and from universities and the private sector. A further key to success will be the substantial commitment of effort that will be obtained from these individuals. Crucial to meeting these objectives will be the ability of TAP to provide the requisite level of funding that can attract outstanding scientists at a high level of commitment. For this reason TAP will concentrate its funding in a rather small number of projects that are sufficiently well funded to meet these requirements.

As Field Studies are the centerpiece of TAP, it is crucial that TAP provide a structure for the effective design, conduct, and interpretation of Field Studies. The TAP approach to this will be by means of Field Study Teams that will constitute a major scientific commitment from a single institution or collaboration of institutions. Members of these teams will be involved in developing and deploying specialized measurements required for the campaigns. The heads of these teams will comprise the membership of the TAP Field Investigators Team that will have responsibility for major scientific direction, such as selection of locations and schedules for TAP Field Campaigns. It is anticipated that each of the different teams will bring a different balance of expertise to the Science Team; for example some teams may bring greater expertise in particle instrumentation, others in gas-phase instrumentation, others in meteorological measurements and their interpretation. Each team will be expected to make a contribution to the science of TAP, including major publications in key scientific journals. The team approach will benefit greatly from the critical mass of effort that can be gained only from a major commitment of several senior investigators, professionals, post-docs and graduate students in a given institution or consortium.

In addition to the Field Study Teams TAP will include several smaller science team projects focusing on a more narrowly defined effort, typically comprising the efforts of a single senior investigator and a post-doc and/or graduate student(s). For modeling activities this effort would be complemented by a

programmer, for which additional funding would be required. Theoretical studies require somewhat lower funding, reflecting a lower fractional effort by the senior investigator as is typical for such studies.

All science team projects will be awarded on the basis of peer-reviewed proposals in response to DOE and/or joint agency announcements.

Estimated funding requirements for the first three years of the Program are given in Table 7.1. During the start-up period of TAP the budget will be ramped up over a three year period. For the Field Study Teams this will be achieved by ramp up of the budgets of the several teams. Other projects will be initiated by program announcements in each of the first three years of the Program. The latter approach will allow targeting of announcements of opportunity to meet specific programmatic requirements as they become identified.

7.2. TAP Science Support Team

The success of a program of the complexity of TAP requires, in addition to the scientific teams and single-investigator projects, a substantial support endeavor. There are numerous activities of science support that are identified in Sections 5 and 6 as necessary for this project. These activities are summarized in Table 7.2 and given in detail in Tables 7.3 and 7.4, along with anticipated budget requirements over the first three years of the project. As discussed in Section 6, the Science Support Team will be populated from groups within the DOE National Labs and elsewhere having the necessary expertise to meet the programmatic requirements.

7.3. TAP Project Support

The TAP Project Support activity (Table 7.5) will be responsible for management of TAP Science Support activities and for all fiscal and contracting aspects of the Program, except for science team funding which will be administered directly by the DOE Program Director.

7.4. Capital Items

For the Science Team components of TAP a capital allocation of 12.5% of operating expenses is indicated in Tables 7.1 and 7.2.

Capital requirements for Science Support instrumentation are given in Tables 7.6 - 7.8 and summarized in Table 7.9. These requirements were evaluated based on expected instrumentation requirements for the supersite, the satellite sites, and the in-situ measurement aircraft.

The success of a program of the complexity of TAP requires, in addition to the scientific teams and single-investigator projects, a substantial support endeavor.

Capital funding in year will be 1 directed to acquisition of instrumentation for the supersite and the in-situ measurement aircraft. Two key instruments will be acquired for each platform, a mobility aerosol analyzer for determining particle size spectrum over the two important decades, 0.01 to 1 μm, and a mass spectrometric instrument that is capable of measuring a broad spectrum of important aerosol precursor gases. The full complement of TAP Science Support instrumentation for the supersite will be acquired in year 1. Acquisition of certain aircraft instrumentation will be deferred to years 2 and 3; this is predicated on the availability of existing instrumentation associated with the DOE Research Aircraft Facility. Instrumentation will be acquired for a single satellite site. This will allow deployment of a satellite site in initial Field Campaigns to evaluate the concept and allow for mid-course corrections.

In Year 2 two more satellite sites will be brought on line. The important addition to the aircraft instrument suite will be three integrating nephelometers, which will allow the relative humidity dependence of the light scattering coefficient to be determined. New particle spectrometers will be acquired to take advantage of recent developments in the state of such instruments.

In Year 3 the final three satellite sites will be brought on line. Upgraded instruments for gas-phase measurements on the research aircraft will also be acquired.

Table 7.1 Overall Summary Budget

Program Element	Year 1 Budget $K	Year 2 Budget $K	Year 3 Budget $K
TAP SCIENCE TEAM			
National Laboratories	3930	7970	12000
Universities and Other Agencies	2840	5475	7850
TAP Science Team Subtotal	*6780*	*13445*	*19850*
TAP SCIENCE SUPPORT	3100	6475	9125
TAP PROJECT SUPPORT	900	900	900
TOTAL OPERATING	**10780**	**20820**	**29875**
Science Team Capital	850	1680	2480
Science Support Capital	1582	642	658
CAPITAL SUBTOTAL	**2432**	**2322**	**3138**
TOTAL OPERATING PLUS CAPITAL	**13212**	**23142**	**33013**

Table 7.2 TAP Science Team (TST) Budget

	Year 1		Year 2		Steady State	
Program Element	Number of Projects	Budget $K	Number of Projects	Budget $K	Number of Projects	Budget $K
National Laboratories						
Lead Scientist.	1	300	1	450	1	600
Field Study Teams	3	2000	3	4000	3	6000
Instrumentation Development and Deployment	2	500	3	750	4	1000
Modeling and Interpretation			1	500	2	1000
Laboratory Studies	2	800	4	1600	6	2400
Theory	1	330	2	670	3	1000
National Laboratories subtotal		***3930***		***7970***		***12000***
Universities, Other Agencies, Private Sector						
Major Field Teams	3	1000	3	2000	3	3000
Instrumentation Development and Deployment	2	500	3	750	4	1000
Modeling and Interpretation	2	450	3	925	4	1400
Laboratory Studies	3	750	6	1500	8	2000
Theory	1	150	2	300	3	450
Universities, Other Agencies, Private Sector subtotal		***2840***		***5475***		***7850***
TAP SCIENCE TEAM TOTAL		***6780***		***13445***		***19850***
Capital for Science Team Projects		850		1680		2480
TAP SCIENCE TEAM TOTAL WITH CAPITAL		**7630**	**0**	**15125**	**0**	**22330**

Table 7.3 TAP Science Support (TSS) Budget

	Budget $K		
Activity per Field Campaign	Year 1	Year 2	Steady State
Planning, Development, Implementation and Interpretation of Major Field Operations	200	500	800
Supersite . Set up, calibrate TSS instruments prior to campaign. Identify site, negotiate occupancy, deploy trailers for TSS and TST instruments, establish power and communications. Site set-up. Installation of PI instruments. Operate TSS instruments during campaigns, deliver preliminary data during campaign, Site break-down, conduct post-operation calibrations as necessary, deliver final data sets, work with TST as required on data interpretation.	500	1400	1800
Satellite sites (4-6). Set up, calibrate TSS instruments prior to campaign. Identify site, negotiate occupancy, deploy instruments, establish power and communications. Site set-up. Operate TSS instruments during campaigns, deliver preliminary data during campaign, Site break-down, conduct post-operation calibrations as necessary, deliver final data sets, work with TST as required on data interpretation.	250	400	600
Meteorological Measurements. Deployment of surface, tower, tether balloon and remote sensing instruments including a network of three Radio Acoustic Sounding System (RASS) wind profilers. Operate instruments during campaigns, deliver data, work with TST in interpretation.	200	450	575
Aircraft operations: Fixed Wing Aircraft for in-situ measurements. Support instrument installations, inlets, Ferry flights, flight hours during 6-week project, delivery of data to Data Assimilation Team. 6 weeks, average of one flight per two days, 5 hours per flight = 105 hours; @$5000 per hour	400	525	525
Aircraft operations: Helicopter for in-situ measurements. As above.	100	300	450
Aircraft operations: Fixed Wing Aircraft for remote sensing and cloud physics measurements.	150	400	525
Instrumentation for airborne measurements: Acquisition, modification for TAP requirements, modifications for flightworthiness, calibration, Operation during campaigns, delivery of preliminary data, De-install from aircraft, post operation calibrations, deliver of final data sets, work with scientists on interpretation of data.	250	300	350
Lidar deployment (e.g., Micropulse Lidar), for continuous characterization of vertical distribution of aerosol at supersite and three satellite sites: Operation, interpretation, providing data to science teams and working with them on use of Lidar data.	50	150	200
Meteorological support: Forecasting, nowcasting, hindcasting. Capturing, and analyzing numerical analysis fields from weather forecast agencies, Execution of mesoscale model such as MM5. Work with science teams in interpretation.	50	100	150

cont'd . . .

Table 7.3 TAP Science Support (TSS) Budget (cont'd)

Tracer deployment and analysis	150	250	500
QC and audits of instruments of TAP investigators. Liaison with QC officers of cooperating entities to establish compatibility of data.	50	100	150
Chemical analysis of major ionic species, elemental and total organic carbon.	150	350	500
Molecular speciation of organics; Analytical costs for supersite only: 50 samples @$5000	200	250	250
TAP Data Management (TDM) See Table 7.4	400	1000	1750
TOTAL	**3100**	**6475**	**9125**

Table 7.4 TAP Data Management (TDM) Budget

	Budget $K		
Activity	Year 1	Year 2	Steady State
Ingest Development @$25K	100	350	600
Level 1,2 product development @50K	100	100	100
Archive		100	300
External Data	50	100	100
Regional Meteorological Model Operations		50	100
Data distribution		100	200
Field Campaign support	125	100	100
Command Center setup		25	25
Field Q/A		25	25
Web server maintenance	25	25	100
Reports		25	100
TOTAL	**400**	**1000**	**1750**

Table 7.5 TAP Project Support Budget

	Budget $K		
Activity	Year 1	Year 2	Steady State
Project Support Director	300	300	300
Fiscal Officer	200	200	200
Contracting	200	200	200
Public Relations and Outreach	100	100	100
Secretarial	100	100	100
TOTAL	900	900	900

Table 7.6 TSS Capital Requirements Super Site

	Budget $K		
Capital Item	Year 1	Year 2	Year 3
SIMS (Secondary Ionization Mass Spectrometer) for selected organics, HNO_3, PAN	450		
Mobility aerosol analyzer for aerosol size distribution 0.01 to 1 µm.	50		
H_2O_2	50		
NO_x	50		
SO_2	6		
O_3	15		
CO	15		
TEOM (Tapered Element Oscillating Microbalance)	10		
Nephelometer	30		
Aerosol absorption monitor	10		
Data system	30		
Pump and Manifold, includes filter housings	10		
Shelter $5K each; four required	20		
TOTAL	**746**	**0**	**0**

Table 7.7 TSS Capital Requirements Satellite Sites

	Budget $K		
Capital Item	Year 1	Year 2	Year 3
Mobility aerosol analyzer for aerosol size distribution 0.01 to 1 µm.	50	100	150
NO_x	10	20	30
SO_2	6	12	18
O_3	15	30	45
CO	15	30	45
TEOM	10	20	30
Nephelometer	30	60	90
Data system	10	20	30
Pump and Manifold, includes filter housings	10	20	30
Shelter	5	10	15
TOTAL	**161**	**322**	**483**

Table 7.8 TSS Capital Requirements Aircraft Instruments

	Budget $K		
Capital Item	Year 1	Year 2	Year 3
SIMS (Secondary Ionization Mass Spectrometer) for selected organics, HNO_3, PAN	450		
Mobility aerosol analyzer for aerosol size distribution 0.01 to 1 μm	160		
NO_x			70
SO_2			20
O_3			25
CO			50
TEOM			10
H_2O_2	50		
Nephelometers (3) w RH control		180	
Particle Spectrometer 0.1-1 μm		70	
Particle Spectrometer 0.3 - 30 μm		70	
Filter sampling system	5		
Miscellaneous, e.g., microscope slide exposure system.	10		
TOTAL	**675**	**320**	**175**

Table 7.9 Total TSS Capital Requirements

	Budget $K		
Platform	Year 1	Year 2	Year 3
Supersite	746		
Satellite sites	**161**	**322**	**483**
Aircraft instrumentation	675	320	175
TOTAL	**1582**	**642**	**658**

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Appendix A

Workshop Participants

After a draft version of this document had been circulated, a workshop was held at Brookhaven National Laboratory June 2-4,1999, to gain input from a broad community representing scientists from DOE National Laboratories, other federal laboratories, universities, and the private sector, and officials in the various federal agencies responsible for air quality and aerosol research. The following individuals participated in this workshop. * Denotes unable to attend in person but provided feedback on the draft report.

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Appendix B

Instrumentation and Characterization Techniques Available for Use in TAP

Instrumentation and measurement techniques that are examples of the state of the art that may be applied in TAP field programs are presented here. Table B1 presents techniques for measurement and characterization of aerosol and single particle size distributions, composition, and other properties. Instrumentation and techniques suitable for characterizing gas-phase species are listed in Table B2. Table B3 lists instrumentation and techniques for characterizing pertinent meteorological variables. The Tables also indicate the suitability of the techniques for use on airborne platforms, the temporal resolution and, for aircraft deployment, the approximate corresponding spatial resolution.

Table B1. Techniques for Characterization of Aerosol Composition and Properties

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
SIZE DISTRIBUTION					
Size Distribution/Particle Formation & Growth	Size and Conc 3-10 nm	Pulse Height Analysis	1-60 sec	+	100 m – 6 km
	Size and Conc 3-150 nm	Mobility Analysis - Nano Differential Mobility Analyzer (DMA)	60 s - 15 min	+	6 km + 9 km
	Size and Conc 3-150 nm	Multiple Condensation Particle Counters at Different Supersaturations	1-60 sec	+	100 m - 6 km
Size Distribution	Size and Conc 10-1000 nm	Mobility Analysis - Long DMA	30 s - 15 min	+	6 km + 9 km
	Size and Conc 1000-2500 nm	Aerodynamic Particle Sizer	5 - 15 min		9 km
Haze and Aerosol Size Distribution	Size and Conc 100-3000 nm	Passive Cavity Aerosol Spectrometer Probe (PCASP)	1-60 sec	+	100 m – 6 km
Cloud and Aerosol Size Distribution	Size and Conc 1-50 µm	Forward Scattering Spectrometer Probe (FSSP)	1-60 sec	+	100 m – 6 km

Cont'd...

Table B1. Techniques for Characterization of Aerosol Composition and Properties, Cont'd.

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
NUMBER, MASS CONCENTRATION					
Total Number Concentration	Conc >3, >10 nm	Ultrafine Condensation Particle Counter (UCPC), Condensation Particle Counter (CPC)	1 - 60 s	+	100 m 6 km
Total Particle Mass	Mass < 1 µm,< 2.5 µm, < 10 µm	Tapered Element Oscillating Microbalance (TEOM)			
	Mass < 1 µm,< 2.5 µm, < 10 µm	Filter collection; humidity controlled weighing	8 h		
CHEMICAL SPECIATION					
Size Distributed Chemical Composition	Anions & Cations 50-10000 nm	Micro-Orifice-Multi-Stage Impactor (MOUDI)	3 hr polluted; 8-24 hr clean		100-1000 km
	Organic compounds 50-10000 nm	Micro-Orifice-Multi-Stage Impactor (MOUDI)	3-24 hr		100-1000 km
Bulk Organic Aerosol Composition	Polar, Non-polar organics, carboxylic acids, 'major' species	Gas Chromatography Mass Spectrometry (GC/MS)	3 hr		
Single Particle Molecular Composition	Elemental Composition 10 nm -1000 nm; possibly size	Time of flight Single Particle Mass Spectrometry (TOF/MS)	> 5 min	+	
Single Particle Electron Microscopy	Size dependent elemental composition down to Carbon - possibly carbon	Transmission electron microscopy	1-10 min	+	6-60 km
Semi-Continuous Bulk Chemical Composition	Particle sulfate and nitrate	Flash vaporization	5-10 min		
Semi-Continuous Water Soluble Chemical Composition	Total mass of major anions and cations for particles	Condensation followed by impaction and collection for Ion Chromatography	5-10 min		
OPTICAL PROPERTIES					
Particle Light Scattering,	Light scattering coefficient for dry particles, one or more wavelengths	Integrating nephelometer	1 s	+	100 m

Cont'd...

Table B1. Techniques for Characterization of Aerosol Composition and Properties, Cont'd.

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
OPTICAL PROPERTIES (cont'd)					
Particle Light Scattering at Elevated Relative Humidity	Light scattering coefficient for particles as f(RH)	RH controlled integrating nephelometer		+	
Particle Sphericity/ Index of Refraction	Light scattering as a function of angular position about levitated particle	Integrating Spherical Scattering Device		?	
Particle Light Absorption	absorption coefficient ~550 nm	Aetholometer	1 min	+	
		Particle Soot Absorption Photometer (PSAP)	1 min	+	
		Photo-acoustic absorption		+	
HUMIDITY RESPONSE AND ACTIVATION					
Particle Water Uptake - function of Relative Humidity	Droplet growth at 20%, 40%, 60%, 80%, 95% RH for 4 Dp's over ambient submicrometer size distribution	Hygroscopic Tandem Differential Mobility Analyzer (HTDMA)	Several Hours	+ (?)	
Cloud Condensation Nuclei (CCN) Spectrum	Concentration of cloud droplets as function of supersaturation	Continuous flow CCN Counter		+	
	Concentration of cloud droplets as function of supersaturation	Thermal Gradient Diffusion Chamber			
REMOTE SENSING					
Aerosol Column Burden	Light extinction	Sun Photometry			
Vertical Profile of Particle Loading	Relative or absolute particle extinction vs. height	Lidar: Depolarization, Backscatter, Raman, and differential absorption (up/down looking)		+	
	Relative or absolute particle extinction vs. height	Satellite (PICASSO-CENA in 2000)			

Cont'd...

Table B1. Techniques for Characterization of Aerosol Composition and Properties, Cont'd.

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
REMOTE SENSING (cont'd)					
Cloud microphysical properties	Radar for microphysical properties of clouds	Ground based; Satellite (NASA Cloudsat in 2003)		+	
Vertical Profile of Particle Size Distribution, Light scatter coefficient	Variation of number concentration with particle size with altitude	Tethered/Free-floating Balloons with aerosol optical probes			

Table B2. Techniques for Characterization of Gas-Phase Species

Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
Hydroxyl radical (OH)	Laser Induced Fluorescence OR ion assisted chemistry/ mass spec OR long path differential absorption	1-60 sec	+	100 m – 6 km
Hydroperoxyl radical (HO_2)	Conversion to OH;Selected Ion Chemical Ionization Mass Spectrometry (SICIMS)			
Ozone (O_3)	Ultraviolet Photometry 2 ppb Detection Limit (DL)	1-60 sec	+	100 m – 6 km
Sulfuric acid (H_2SO_4)	SICIMS or Trace Atmospheric Gas Analyzer (TAGA) 1.5 ppt DL	1-60 sec	+	100 m - 6 km
Ammonia (NH_3)	TAGA/500 ppt DL or Atmospheric Pressure Chemical Ionization Mass Spectrometer (API-MS)	1-60 sec	+	100 m – 6 km
Nitric acid (HNO_3)	SICIMS 10 ppt; TAGA 100 ppt DL	1-60 sec	+	100 m – 6 km
Nitric oxide (NO)	Chemiluminescence 10 ppt DL	1-30 sec	+	100 m – 6 km
Nitrogen dioxide (NO_2)	UV-Photolysis followed by chemiluminescence 50 ppt DL	4-30 sec	+	400 m 6 km
Total NO_y	Molybdenum catalyst; chemiluminescence 50 ppt DL	1-60 sec	+	100 m – 6 km
Sulfur dioxide (SO_2)	Pulsed Fluorescence 200 ppt DL also TAGA/100 ppt DL	15-60 sec	+	1.5 – 6 km
Carbon monoxide (CO)	Gas-filter correlation 15 ppb DL or long path vacuum UV absorption (DACOM) or reducing gas detector	30-60 sec	+	3-6 km

Cont'd...

Table B2. Techniques for Characterization of Gas-Phase Species, Cont'd.

Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
Formaldehyde (HCHO	Derivatization; High Pressure Liquid Chromatography (HPLC) or Tunable Diode Laser	20-120 s		
Hydrogen peroxide (H_2O_2)	Glass Scrubber Selective Derivatization Fluorimetry 60 ppt DL	60 sec	+	6 km
Hydrocarbons	Canister/Capillary Gas Chromatograph 10-100 ppt DL	30-90 s	+	
Carbonyls	Glass Scrubber High Pressure Liquid Chromatography 10-20 ppt	180 sec	+	18 km
Dimethylsulfide (DMS)	TAGA 2 ppt DL	20-60 sec	+	2-6 km
Nitrous acid (HNO_2)	TAGA 500 ppt DL	20-60 sec	+	2-6 km
Formic acid; acetic acid (HCOOH, CH_3COOH	TAGA 25 to 30 ppt DL	20-60 sec	+	2-6 km
PeroxyacetylInitrate (PAN)	Need 0.05-70 ppb			
Nitrate radical (NO_3)	Need 5-430 ppt			
Polycyclic Aromatic Hydrocarbons				
Radon				

Table B3. Techniques for Characterization of Meteorological Processes

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
Real-Time Air Parcel Trajectory	Air parcel x,y,z position with time	Smart Balloon/ Perfluorocarbon or Tracer Tagging	1-60 sec	+	100 m – 6 km
Boundary Layer Mixing Height	Boundary Layer Height	Lidar	1-60 sec	+	100 m – 6 km
Boundary Layer Mixing and Stratification	Vertical Profile of Temperature, RH, wind speed, wind direction	Radio Acoustic Sounding System/ Rawinsonde/ Tethered Balloon	1-60 sec	+	100 m - 6 km
Entrainment at Top of Boundary Layer	Water Vapor or Ozone	Fast water vapor or ozone concentrations at BL top			100 m – 6 km
Vertical Profile of Winds	Horizontal wind speed and direction with altitude	Sodar			
Local Source Region	Surface Horizontal Wind Speed and Direction	Anemometer	1 sec	+	100 m
Air Mass Thermodynamic State	Temperature, Dew Point Temperature, Pressure	Various	1 sec	**+**	100 m
Boundary Layer Height; Vertical Profile of BL Thermodynamic State	Temperature, Dew Point Temperature, Wind Speed, Wind Direction as a function of Pressure	Rawinsonde	4 hr	**+**	
		Tethered Balloon	Continuous		
		Aircraft Vertical Profiles	15 min		
Precipitation	Volume of precipitation over known time	Tipping Bucket	0.01” precip	**NO**	
Synoptic Scale Forcing	National Weather Service Standard Synoptic pressure analyses – variation of geopotential height along isobars	NWS analysis of surface and upper air pressure fields using rawinsonde data	6 hr	**NO**	
Cloud Processing and Precipitation Along Back Trajectory	Cloud areal coverage, altitude, thickness and precipitation from radar reflectance	Regional NEXRAD images	~ minutes	**NO**	

Cont'd...

Table B3. Techniques for Characterization of Meteorological Processes, Cont'd.

Measurement	Measurand	Technique	Time Resolution	Airborne Platform	Spatial Resolution
Model Predicted Air Mass Back Trajectory	Air mass x, y, and z position with time	Model	One trajectory/ hour	**NO**	Each major sampling site
Synoptic scale Forcing/Free Troposphere – Troposphere Exchange; Cloud Venting	Visible and IR reflectance	Hemispheric and Regional NWS and other Satellites	~Hours		~50-100's km
Cloud Venting and Subsidence; Cloud Top Entrainment; Cloud Bottom In-Flow	Flux of water vapor out of top and possibly sides of cloud; downward flux around cloud	Above cloud eddy correlation using aircraft gust probe and fast response water vapor	5 ms	**?**	0.5 m
Cloud Microphysics; cloud supersaturation	Cloud Updraft Velocity	Gust Probe	5 ms	**+**	0.5 m
Photochemical Potential	Downwelling Solar Radiance			**+**	
Vertical Temperature Profile; Cirrus Cloud Properties	Upwelling Infrared Radiance; Cirrus thickness, transmissivity and areal coverage	Satellite IR sensor			

Appendix C

Relationship between TAP and other Federally Funded PM Research Programs

A number of related research programs directed to research on atmospheric particulate matter are currently conducted by various Federal agencies. According to the recently published Inventory of Federal Research Programs prepared by the Air Quality Research Subcommittee (AQRS, 1998) of the Committee on the Environment and Natural Resources, approximately $26M was devoted to process research pertinent to aerosol formation and $4M to exposure assessment and risk analysis. The distribution of resources among the PM process-related research is shown in Figure C1.

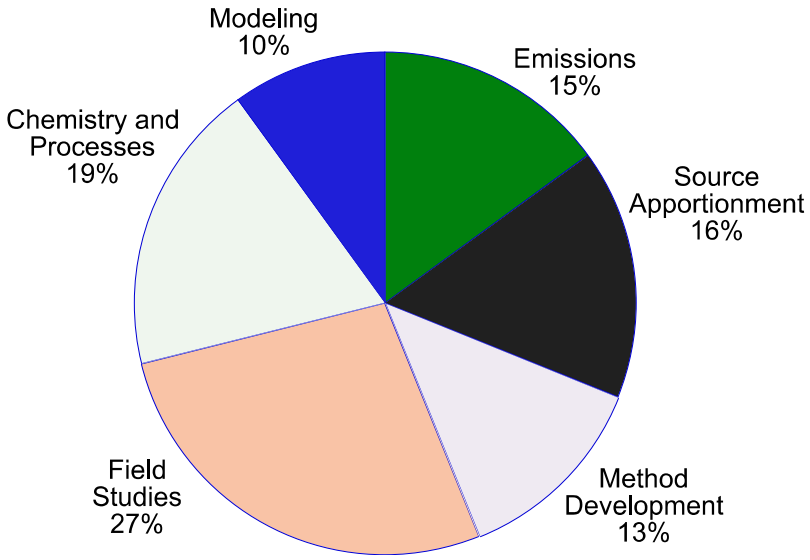

Figure C1. Allocation of resources within atmospheric process research on particulate matter in the Federal sector. Adapted from AQRS(1998).

Much complementary research is conducted under the aegis of global change research (SGCR, 2000). The various projects and respective funding agencies are presented in Table C1 categorized according to the project research focus.

Table C1. Compilation of Federal Agency Research Projects on atmospheric Particulate Matter, grouped according to research focus. Adapted from AQRS(1998).

Component	Program/Effort	Agency	$, M 1998	Geographical scale
Methods and Monitoring	IMPROVE	NPS/DOI	1.24	National
	AIRMoN, chemical deposition by particles	NOAA	0.2	National
	ARM/PM	DOE/OBER	1.0	National
	PM2.5 Sampling/Analysis	DOE/NETL	1.5	Ohio River Valley
	Southern Oxidants Study PM	EPA/ORD	0.4	Southeast
	New PM Analytical Methods	EPA/ORD	1.0	
	Exploratory Grants on PM Environmental characterization and measurement Method	EPA/ORD	2.1	
	FRM and Equivalency program for PM2.5	EPA/ORD	0.37	
	FRM Monitoring Partnerships and Regional Supersite	TVA	0.23	Tennessee Valley region
	PM Remote Sensing	NASA		Global
Chemistry, Meteorology, and Modeling	Processes of fine PM formation and distribution	NOAA, Air Quality	0.45	National
	Observations and Modeling	NOAA, Global Change	0.95	National
	Processes and Distribution	NASA /GACP	2.0	Global
	ACP/Aerosol/Indirect	DOE/OBER	2.0/4.0	National
	Processes affecting pest control application	USDA		
	Formation, Fate, and Composition of Tropospheric PM	NSF	1.5	Regional, Global
	Organic Aerosols Chemistry for Multi-Scale Modeling	EPA/ORD	0.5	National
	Models-3/CMAQ for PM	EPA/ORD	1.7	National
	EMP-indirect	DOE/OBER	2.0	National

Table C1, cont'd. Compilation of Federal Agency Research Projects on atmospheric Particulate Matter, grouped according to research focus. Adapted from AQRS(1998).

Component	Program/Effort	Agency	$, M 1998	Geographical scale
Chemistry, Meteorology, and Modeling (cont'd)	Exploratory grants on PM atmospheric chemistry and modeling	EPA/ORD	1.15	
Emission Characterization & Source Apportionment	BRAVO	NPS/DOI	0.9	Big Bend National Park
	Origins and Dispersion of Primary PM	NOAA/DOC	0.2	National
	Smoke Management and Air Quality	USDA	1.9	National
	Wind Erosion and Air Quality Prediction	USDA	0.8	WA
	Emissions from Cotton Ginning	USDA	0.05	TX, OK, NM
	Receptor Modeling and Source Chemical Profiles	EPA/ORD	0.8	National
	Source Apportionment and CMB Analysis of Urban Sources	EPA/ORD	0.19	National
	PM Emissions from Indoor and Outdoor Sources	EPA/ORD	1.6	National
	Source, Formation, and Transport in the Tennessee Valley	TVA	0.5	Tennessee Valley region

A major portion of programs that are identified as field studies are carried out primarily for monitoring purposes, e.g., the IMPROVE program, as distinguished from research directed to aerosol evolution in the atmosphere. Only limited research effort is directed to gaining a quantitative understanding of the life cycle of aerosols in a geographical scale of ~200 km based on a detailed field measurements involving a comprehensive suite of chemical and meteorological measurement platforms. TAP is complementary to many ongoing projects in a number of ways. For example, method and model development are an integral part of the TAP effort, and emission characterization and source apportionment will in some instances serve as model inputs and in others be tested by field observation data.

Reference

AQRS (1998). Air Quality Research Subcommittee, Committee on Environment and Natural Resources. Atmospheric Particular Matter Research: Inventory of Federal Research Programs. September, 1998.

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Appendix D

Acronyms

ACP	DOE Atmospheric Chemistry Program
ACPI	DOE Accelerated Climate Prediction Initiative
AQRS	Federal Interagency Air Quality Research Subcommittee
ARM	DOE Atmospheric Radiation Measurement Program
BERAC	DOE Biological and Environmental Research Advisory Committee
BL	Boundary Layer
CARB	California Air Resources Board
CMAQ	Community Multiscale Air Quality model
CMB	Chemical Mass Balance
DL	Detection Limit
DOC	Department of Commerce
DOE	Department of Energy
DOI	Department of Interior
EC	Elemental Carbon
EMP	DOE Environmental Meteorology Program
EMSL	Environmental Molecular Sciences Laboratory
EOS	NASA Earth Observing System
EPA	Environmental Protection Agency
ESD	Environmental Sciences Division within DOE/OBER
FRM	Federal Reference Method
GCRS	Federal Interagency Global Change Research Subcommittee
HEI	Health Effects Institute
IDAC	Instrument Development and Advanced Characterization component of TAP
IPCC	Intergovernmental Panel on Climate Change
NAAQS	National Ambient Air Quality Standards
NAPAP	National Acid Precipitation Assessment Program
NARSTO	formerly North American Research Strategy on Tropospheric Ozone
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NETL	National Energy Technology Laboratory
NOAA	National Atmospheric and Oceanic Administration
NPS	National Park Service
NRC	National Research Council
NSF	National Science Foundation
OBER	DOE Office of Biological and Environmental Research
OC	Organic Carbon
PM2.5	Particulate Matter of aerodynamic diameter greater than or equal to 2.5 μm
PM10	Particulate Matter of aerodynamic diameter greater than or equal to 10 μm
PNNL	Pacific Northwest National Laboratory
QA/QC	Quality Assurance/Quality Control
RASS	Radio Acoustic Sounding System
SGCR	Federal Subcommittee on Global Change Research
SICIMS	Selected Ion Chemical Ionization Mass Spectrometry
SIMS	Secondary Ionization Mass Spectrometer
SOS	Southern Oxidants Study
SSC	Scientific Steering Committee, of TAP
TAGA	Trace Atmospheric Gas Analyzer

TAP	Tropospheric Aerosol Program
TDM	TAP Data Management
TEM	Transmission Electron Microscopy
TEOM	Tapered Element Oscillating Microbalance
TOF-MS	Time-Of-Flight Mass Spectrometry
TSS	TAP Science Support
TST	TAP Science Team
TVA	Tennessee Valley Authority
USDA	United States Department of Agriculture
USGCRP	U.S. Global Change Research Program
UTC	Universal Time Coordinated
VOC	volatile organic compound
VTMX	Vertical Transport and Mixing Program within DOE/OBER/ESD